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Scheme of a hydrogen-molecule quantum simulator based on two ultracold rubidium atoms

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Abstract. **A scheme is proposed for implementing a hydrogen-molecule quantum simulator based on two ultracold rubidium atoms trapped into spatially separated optical dipole traps. The scheme includes the adiabatic preparation of the initial quantum state of two atoms and the iterative quantum phase estimation. The accuracy of measuring the ground state energy of a molecule is numerically calculated as a function of the number of iterations. The simulation is performed using two-qubit gates based on the dipole blockade effect under short-term excitation of atoms into the Rydberg states with allowance for the finite lifetime of Rydberg states and the finite energies of the van der Waals interaction.**

Keywords: *ultracold rubidium atoms, hydrogen molecules, quantum simulator.*

1. Introduction

Quantum simulation of complex physical systems is one of the most interesting problems of modern physics [1, 2]. The main problem of such a simulation on a classical computer is the exponential growth of the required computational resources with an increase in the complexity of the simulated system. In this regard, in recent decades, methods for the implementation of quantum simulators have been actively developing.

There are two types of quantum simulators [3]. The so-called analogue quantum simulators are artificial quantum systems consisting of separate quantum objects. The interaction between them is described by the operators similar to the interaction operators in more complex physical systems. The main difficulties in designing such simulators are associated with the need to find suitable artificial systems and select their parameters, which would be feasible for describing real conditions of an experiment. Another type is the so-called digital quantum simulators similar to quantum computers. In such simulators, algorithms for estimating the phase of an arbitrary unitary matrix are used, allowing one to find its eigenvalues. If a propagator for any physical system is chosen as a unitary operator, it is possible to find the energy eigenvalues of the corresponding Hamiltonian.

The advantages of digital quantum simulators consist in their versatility, and also in the wide possibilities of their application, in particular, in the field of quantum chemistry [4]. The main problem of quantum chemical calculations is to find the possible structure and properties of individual molecules or molecular formations. This problem can be reduced to the search for energies of various states of a molecule, or, in other words, to the search for eigenvalues of a certain unitary operator. In particular, the known-to-date algorithms for finding the molecule eigenstates are based on the phase estimation algorithm (PEA) proposed by Abrams and Lloyd [5].

The main difficulties in the development of digital quantum simulators are primarily related to the limited accuracy of the two-qubit gates required for quantum phase estimation. In addition, the decomposition of unitary operators of arbitrary dimension, which is necessary for simulation of complex multi-particle systems, is a nontrivial mathematical problem [2].

One of the promising approaches to the implementation of quantum simulators is the use of cold atoms in arrays of optical dipole traps. Such arrays can be used to design both analogue and digital quantum simulators, which were described in detail in [3, 6]. A quantum simulator consisting of 51 cold rubidium atoms in a one-dimensional array of optical dipole traps was demonstrated in [7]. Examples of modern analogue simulators and their possible applications were discussed in detail in [8]. At the same time, the problem of low accuracy of the two-qubit gates remains relevant for atomic systems, which complicates the implementation of digital quantum simulators. In this regard, of great interest at this stage is the implementation of a simplest digital quantum simulator that can be developed on the basis of two neutral atoms trapped in spatially separated optical dipole traps.

2. Algorithms of quantum phase estimation

The simplest example of the implementation of a digital quantum simulator is the determination of the binding energy of the ground state of the hydrogen molecule. Du et al. [9] solved experimentally this problem by the Aspuru-Guzik method [10] with the use of nuclear magnetic resonance. To find the solution to this problem, it is sufficient to use two

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qubits: the state of one of them encodes the ground state of hydrogen molecule, while the second qubit serves to measure the phase. The authors experimentally used the adiabatic state preparation (ASP) method and iteratively measured the phase, thereby receiving 45 significant bits of data.

Following work [9], we used an STO-3G minimal basic set for the 1s orbital [11]. At a distance of 1.4 a.u. between the nuclei of atoms in the hydrogen molecule, the Hamiltonian matrix (in atomic units) has the form [9]:

$$\hat{H}_{\text{mol}} = \begin{pmatrix} -1.8310 & 0.1813 \\ 0.1813 & -0.2537 \end{pmatrix}. \quad (1)$$

As shown in Fig. 1a, the phase estimation algorithm [5, 10] uses two quantum registers: the state register S, into which the wave function of the system state $|\psi\rangle$ is being written, and the readout register R needed for storing intermediate information and obtaining the eigenvalues of the energy E. We denote the states of the register R as $|n\rangle$. The Hamiltonian H_{mol} of the system in question is used to generate a unitary evolution operator

$$\hat{U}|\psi\rangle = \exp(-\mathrm{i}\hat{H}_{\text{mol}}\tau)|\psi\rangle = \exp(2\pi\mathrm{i}\varphi)|\psi\rangle. \quad (2)$$

By measuring the phase φ, one can find the eigenvalue of the Hamiltonian (the system energy) $E_{\text{meas}} = -2\pi\varphi/\tau$. To this end, we sequentially, in a controlled manner, apply the operators $\hat{U}_k = \exp(-\mathrm{i}\hat{H}_{\text{mol}}\tau)^{2^k}$ to the register S and ultimately obtain the following system state:

$$|R\rangle\otimes|S\rangle = \sum_n \exp(2\pi\mathrm{i}\varphi n)|n\rangle\otimes|\psi\rangle. \quad (3)$$

Then we perform the inverse quantum Fourier transform with the register R and measure its state. This allows us to evaluate the phase with arbitrary accuracy. Note that the register state is measured only once.

This algorithm can be implemented even if the register R consists of a single qubit (Kitaev's phase estimation algorithm, Fig. 1b) [2, 12]. After a controlled unitary transformation, the two-qubit system passes to the state $(1/\sqrt{2})[|0\rangle + \exp(2\pi\mathrm{i}\varphi)|1\rangle]|\psi\rangle$. The system after the Hadamard gate is in the state

$$\frac{1}{2}[1 + \exp(2\pi\mathrm{i}\varphi)]|0\rangle|\psi\rangle + \frac{1}{2}[1 - \exp(2\pi\mathrm{i}\varphi)]|1\rangle|\psi\rangle. \quad (4)$$

The probability of finding the controlled qubit in the state '0' is $P = \cos^2(\pi\varphi)$. By measuring the probability P, one can find the phase φ. Instead of a single measurement of the state of a multi-qubit register, multiple measurements of the state of a single qubit are required to attain the prescribed accuracy.

A distinctive feature of the Aspuru-Guzik algorithm [10] shown in Fig. 1c is its recursiveness, allowing one to signifi-

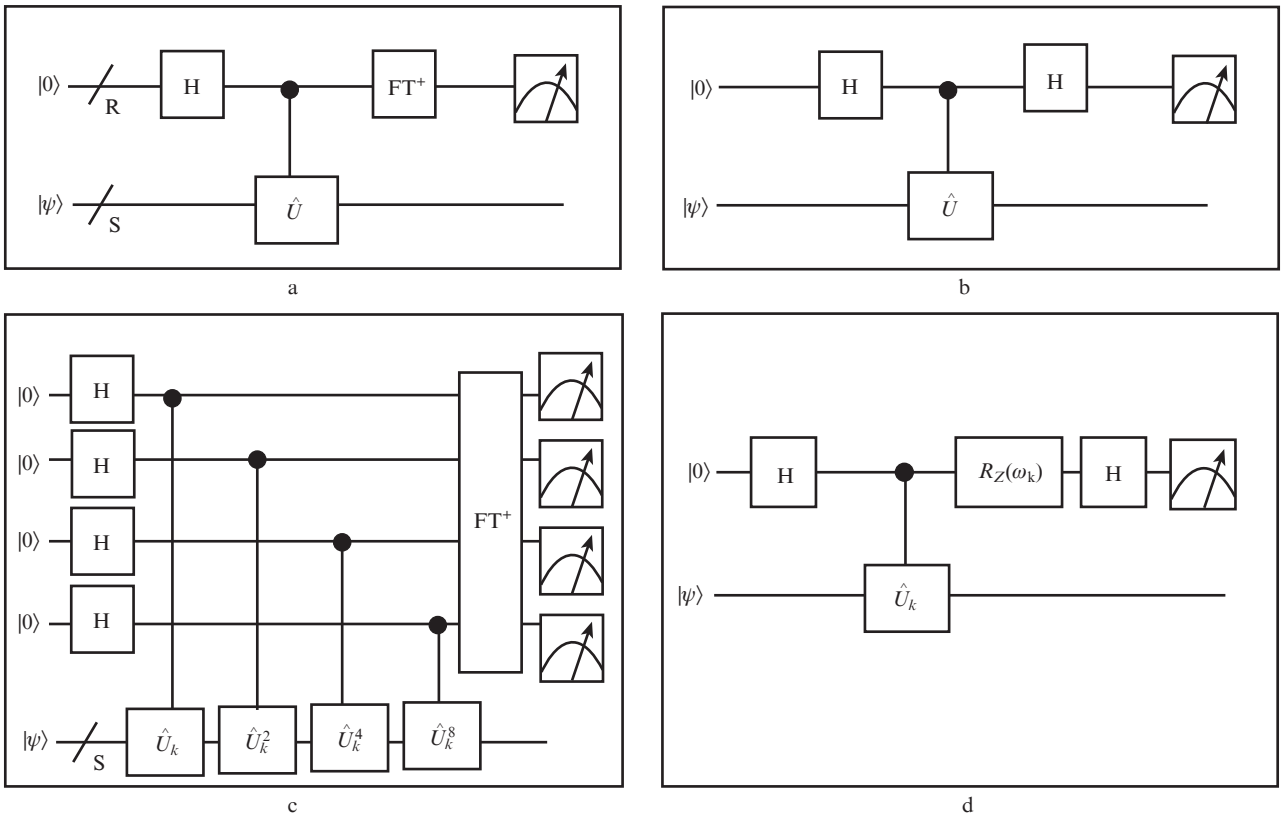

Figure 1. (a) General scheme of quantum phase estimation, (b) Kitaev's quantum phase estimation algorithm [2, 12] using a single controlling qubit, (c) Aspuru-Guzik phase estimation algorithm [10], and (d) adaptive phase estimation scheme using a single measurement of the controlling qubit at each iteration step [13]. Here H is the Hadamard gate; $\hat{U} = \exp(-\mathrm{i}\hat{H}_{\text{mol}}\tau)$; $\hat{U}_k = \exp(-\mathrm{i}\hat{H}_{\text{mol}}\tau)^{2^k}$; FT^+ is inverse quantum Fourier transform; and $R_Z(\omega_k)$ is the qubit rotation around the Z axis by the angle ω_k.

cantly reduce the number of qubits in the register R, while attaining the required accuracy in phase estimation. In original paper [10], it was proposed to use four qubits in register R, as shown in Fig. 1c. In the first step, the operator $\hat{U}_0 = \hat{U}$ is applied to the register S. This allows one to estimate the phase φ with an accuracy of 15/16. After that, a unitary transformation

$$\hat{U}_k = [\exp(-2\pi i\varphi'_{k-1})\,\hat{U}_{k-1}]^2 \qquad (5)$$

is performed at each iteration step. Here φ'_{k-1} is the lower boundary of the phase estimate obtained at the previous step. Thus, at each subsequent iteration step, the next significant bit is measured. Du et al. [9] used the Aspuru-Guzik algorithm for the register R consisting of one qubit. In their work, they measured three bits at each iteration step, with each iteration step leading to an increase in the accuracy by one bit.

An interesting method of iterative phase estimation was proposed in [13]. Its advantage is that each next significant bit can be obtained as a result of a single measurement of the state of an auxiliary qubit. The phase estimation scheme is shown in Fig. 1d. In contrast to the above-considered schemes, the measurement starts from the low-order bit of the phase. Each iteration includes a single measurement of the state of the controlling qubit. Before the measurement, its state is adjusted by means of the phase shift $R_Z(\omega_k)$ in order to take into account the results of previous measurements.

In accordance with the notation adopted in the monograph by Nielsen and Chuang [2], we represent the phase φ as a sequence of m bits, $\varphi_1 ... \varphi_m$, in the form: $\varphi = 0.\varphi_1\varphi_2 ... \varphi_m = \varphi_1/2 + \varphi_2/4 + ... + \varphi_m/2^m$. At the first iteration of the algorithm, a controlled transformation $\hat{U}_m = \exp(-i\hat{H}_{\rm mol}\tau)^{2^m}$ is performed (here we put $\omega_m = 0$). The probability of obtaining the '0' result when measuring the state of the first qubit is $P_0 = \cos^2[\pi(0.\varphi_m)]$, since all the high-order bits of the phase do not affect the result of measurements due to periodicity of the complex-valued exponent. Since φ_m takes only two possible values, it can be measured only once.

The next step is the controlled rotation, $\hat{U}_m = \exp(-i\hat{H})^{2^{m-1}}$. The system state after its implementation, due to periodicity of the complex-valued exponent, can be represented as follows: $(1/\sqrt{2})[|0\rangle + \exp(2\pi i 0.\varphi_{m-1}\varphi_m)\,|1\rangle]\,|\psi\rangle$. In order to exclude the already measured bit φ_m from the problem we perform rotation of the control qubit $R_Z(\omega_{m-1})$ by the angle $\omega_{m-1} = -2\pi(0.0\varphi_m)$, as shown in Fig. 1d. As a result, after performing the Hadamard gate, the probability of finding the controlling qubit in the '0' state is $P_0 = \cos^2[\pi(0.\varphi_{m-1})]$, which allows us to determine the value of the bit φ_{m-1} as a result of a single measurement.

All subsequent bits are measured in the same way. As shown in [13], the accuracy of this algorithm is equal to the accuracy of the standard measurement based on the inverse quantum Fourier transform of the register consisting of m bits.

3. Adiabatic preparation of the initial qubit state

Quantum phase estimation algorithms require the controlled qubit to be initially prepared in the eigenstate $|\psi\rangle$ of the Hamiltonian $\hat{H}_{\rm mol}$. To this end, the method of adiabatic preparation of initial state can be used [9, 10]. Initially, the qubit is prepared in the state $(1/\sqrt{2})(|0\rangle - |1\rangle)$, which is an eigenstate for the matrix

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}.$$

Let the auxiliary Hamiltonian

$$\hat{H}_{\rm ad}(t) = \left(1 - \frac{t}{T}\right)\sigma_x + \frac{t}{T}\hat{H}_{\rm mol} \qquad (6)$$

describe a slow evolution of the system Hamiltonian from σ_x to $\hat{H}_{\rm mol}$ during the time T expressed in dimensionless relative units. If the system dynamics is described by the evolution operator $\hat{U}\,|\psi\rangle = \exp(-i\hat{H}_{\rm ad}t)\,|\psi\rangle$, the qubit will remain in the eigenstate of the Hamiltonian $\hat{H}_{\rm ad}$ and ultimately find itself in the eigenstate of the Hamiltonian $\hat{H}_{\rm mol}$. The evolution of the system state during the time T can be divided into M steps. The transformation of the qubit state at each step m is described by the evolution operator

$$\hat{U}^m_{\rm ad}(t) = \exp\left[-i\left(1 - \frac{m}{M}\right)\frac{\Delta T}{2}\sigma_x\right]\exp\left[-i\frac{m\Delta T}{M}\hat{H}_{\rm mol}\right]$$

$$\times \exp\left[-i\left(1 - \frac{m}{M}\right)\frac{\Delta T}{2}\sigma_x\right]. \qquad (7)$$

Here $\Delta T = T/M$, and m takes values from 1 to M. The evolution operator can be represented as a sequence of single-qubit rotations by using the Trotter formula [14]

$$\exp[t(\hat{A} + \hat{B})] = \lim_{n\to\infty}\left[\exp\left(\hat{A}\frac{t}{n}\right)\exp\left(\hat{B}\frac{t}{n}\right)\right]^n, \qquad (8)$$

and the basis expansion $\hat{H}_{\rm mol} = {}^1/_2 \sum_i {\rm Tr}(\hat{H}_{\rm mol}\sigma_i)\,\sigma_i$, where $i = 0, x, y, z$.

The accuracy of initial state preparation can be defined as $|\langle\psi_{\rm ad}|\psi\rangle|^2$, where $|\psi_{\rm ad}\rangle$ is the state obtained as a result of adiabatic preparation. The numerically calculated dependence of the accuracy on the number of steps at $T = 18.4$ and $M = 51$ is shown in Fig. 2. The numerical error did not exceed 10^{-2}.

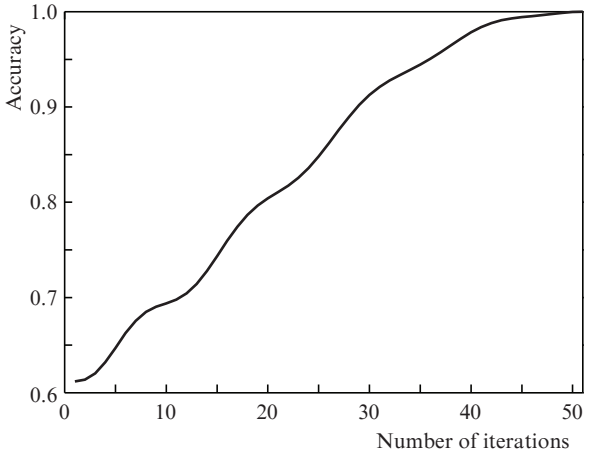

Figure 2. Numerically calculated dependence of initial state accuracy of a qubit on the number of iterations in the case of adiabatic state preparation.

4. Scheme of quantum simulation using ultracold neutral rubidium atoms

In quantum simulation of the hydrogen molecule, we consider two rubidium atoms trapped in spatially separated optical dipole traps formed by tightly focused laser beams [15]. The long-lived superfine sublevels of the ground state of atoms are used as the logical states '0' and '1' of qubits. Single-qubit gates with ultracold neutral atoms can be implemented using microwave radiation or two-photon optical Raman transitions between the superfine sublevels of the ground state of atoms. Such gates were demonstrated with high accuracy in [16, 17].

The interaction of a two-level system with resonant laser radiation can be described by the matrix [18]:

$$R(\theta,\chi)=\begin{pmatrix}\cos\frac{\theta}{2} & \mathrm{i}e^{-\mathrm{i}\chi}\sin\frac{\theta}{2}\\ \mathrm{i}e^{\mathrm{i}\chi}\sin\frac{\theta}{2} & \cos\frac{\theta}{2}\end{pmatrix}. \quad (9)$$

Here, the angle θ is given by the laser pulse area, and the angle χ is given by its phase. The values $\chi=0$ and $\chi=\pi/2$ correspond to the qubit rotations by the angle θ around the X and Y axes, respectively. All single-qubit gates required for adiabatic preparation of the initial state and transformation of the states of individual qubits during the iterative phase estimation can be reduced to a sequence of X- and Y-rotations.

The main difficulty is to implement controlled two-qubit unitary operations. To this end, the scheme shown in Fig. 3, similar to that proposed by us in work [18–20], can be used. In that scheme, the dipole blockade effect is employed, which consists in the impossibility of simultaneous excitation of closely spaced atoms into the Rydberg states [21].

If the controlling atom is initially in the '1' state and is not excited by a laser π-pulse (*1*) to the Rydberg state $|r\rangle$, the result of a sequence of laser π-pulses (2–6) is the transformation described by matrix (9). If the controlling atom is initially in the '0' state, it is excited to the state $|r\rangle$ with the laser pulse *1*. Due to dipole blockade which excludes the excitation of both atoms to collective states $|rr'\rangle$ or $|r'r\rangle$, the action of pulses *2*–*6* is ineffective. Then the controlling atom returns to its initial state under the action of laser pulse *7*. In order to exclude an undesirable phase shift during sequential excitation and de-excitation of Rydberg states, the sign of Rabi frequencies of pulses *5*, *6*, *7* is opposite compared to that of pulses *1*, *2*, *3*. This corresponds to the phase shift of laser radiation by π, as shown in Fig. 3.

Experimental implementation of this scheme requires the choice of states $|r\rangle$, $|r'\rangle$, $|r''\rangle$ strongly interacting with the simultaneous excitation of two atoms to collective states $|rr'\rangle$ and $|rr''\rangle$. As an example, consider the states $|r=81\mathrm{S}\rangle$, $|r'=80\mathrm{S}\rangle$, and $|r''=82\mathrm{S}\rangle$. The main contribution to the interaction of atoms in the states $|81\mathrm{S},80\mathrm{S}\rangle$ is made by Förster resonance $|81\mathrm{S},80\mathrm{S}\rangle \rightarrow |80\mathrm{P}_{3/2},80\mathrm{P}_{3/2}\rangle$ with an energy defect of –161 MHz [22]. The van der Waals interaction energy for this channel is $U=4750$ GHz μm^6. The main contribution to the interaction of atoms in the states $|81\mathrm{S},82\mathrm{S}\rangle$ is made by Förster resonance $|81\mathrm{S},82\mathrm{S}\rangle \rightarrow |80\mathrm{P}_{3/2},81\mathrm{P}_{3/2}\rangle$ with an energy defect of 110 MHz. The van der Waals interaction energy for this channel is $U=-7350$ GHz μm^6. Thus, the energy shift impeding laser excitation exceeds 100 MHz already at an interatomic distance of 4 μm or less. The energy gap between the 80S and 82S states constitutes 27.9 GHz. Two-photon transitions between these states can be implemented using microwave radiation which does not affect the controlling atom in the 81S state.

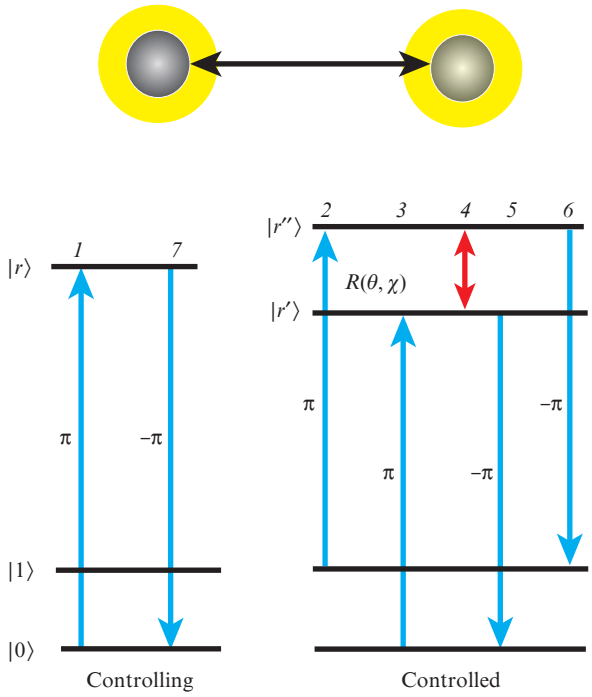

Figure 3. Scheme of controlled qubit rotation for two ultracold atoms in optical dipole traps. Laser π-pulses *1*, *2*, *3*, *5*, *6*, and *7* excite and de-excite Rydberg states with individual addressing to particular atoms. Microwave pulse *4* has an arbitrary area and phase.

For numerical simulation of this experiment, we decomposed the unitary transformation $\hat{U}_m=\exp(-\mathrm{i}\hat{H}\tau)^{2^{m-1}}$ as a sequence of X- and Y-rotations by the given angles calculated separately for each iteration step. In the calculations, we took into account the finite lifetime of the Rydberg states 80S (τ = 208 μs), 81S (τ = 214 μs), and 82S (τ = 221 μs) at a temperature of 300 K [23] and the finite energies of the van der Waals interaction at a distance of 8 μm between the optical dipole traps. To simulate the measurement process, we calculated the probability of finding the controlling qubit in the '0' state and took its state for '0' if the obtained probability exceeded 0.5. Note that for all bits except the low-order one, the numerically calculated probability is close either to 0 or 1 for each iteration step.

Figure 4 shows the numerically calculated dependence of the relative error $|(E-E_{\mathrm{meas}})|/E$ in determining the binding energy of the ground state of the hydrogen molecule on the number of iterations of the phase estimation algorithm. Here E is the eigenvalue of the matrix $\hat{H}_{\mathrm{mol}}$, and E_{meas} is the result of numerical simulation using the quantum algorithm. The step-wise nature of the dependence is due to the fact that the accuracy changes only when the next measured significant bit turns out equal to 1. Accounting for the finite lifetime of Rydberg states and finite energy of the van der Waals interaction does not lead, at the chosen experimental parameters, to the deterioration of the calculation accuracy which is only determined by the number of iterations. Our calculations show that the relative error does not exceed 10^{-14} at 50 iterations. We neglect the finite accuracy of one-qubit gates, and also the effect of the finite width of the laser radiation line, finite lifetime of intermediate excited states (when using, for example, two-photon laser excitation of

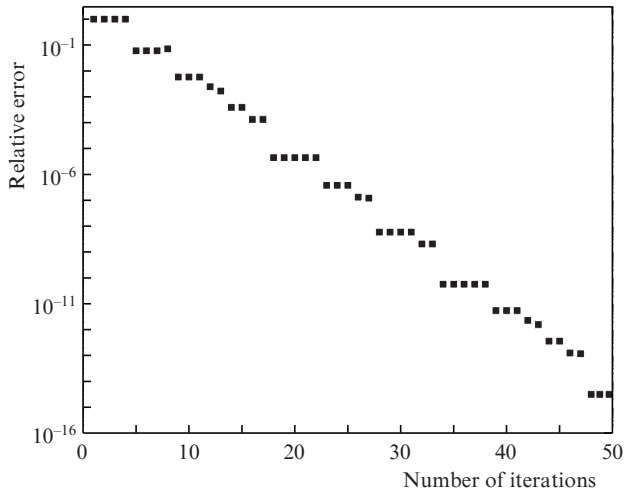

Figure 4. Numerically calculated dependence of the relative error of measuring the ground state energy of the hydrogen molecule on the number of iterations in the case of the phase estimation algorithm.

Rydberg states), motion of atoms, and parasitic electric and magnetic fields.

An interesting feature of the results obtained is that the algorithm sensitivity to errors in performing the two-qubit rotations has a threshold character. This is due to the discreteness of the measured qubit state at each iteration step. If the probability of finding a qubit in the state '0' falls into the interval between 0.5 and 1, in the case of a single measurement it can be detected in the state '1', which leads to a computational error. The finite lifetime of Rydberg atoms and finite energies of the van der Waals interaction lead to a decrease in the probability of a correct measurement of the qubit state. In our calculations, at a distance of 8 μm between the atoms, the probability of erroneous measurement of the state of a single bit (except for the three low-order bits) does not exceed 2%. These errors can be easily eliminated at the expense of several additional measurements at each iteration step.

5. Conclusions

We have proposed a scheme for the implementation of a quantum simulator of the hydrogen molecule using two ultracold atoms in optical traps. The scheme is based on the adiabatic state preparation and iterative quantum phase estimation with a single measurement of the final state of the atom at each iteration step. The advantage of this iterative method is that each iteration step requires a single measurement of the quantum state of a controlling qubit. The accuracy of measuring the ground state energy of the molecule is numerically calculated as a function of the number of iterations. In our simulation, the adiabatic preparation of the initial state is presented as a sequence of one-qubit rotations around the X and Y axes. To implement the quantum phase estimation algorithm, we have proposed a scheme for performing controlled rotations using the dipole blockade effect under the excitation of two atoms to Rydberg states. The calculations show that accounting for the finite lifetime of Rydberg levels and the final energies of the van der Waals interaction makes it possible to attain a high accuracy of the molecule energy measurement (the relative error does not exceed 10^{-14} at 50 iteration steps).

Experimental implementation of this algorithm is of interest for demonstrating the simplest digital quantum simulator based on ultracold neutral atoms. The most significant limitation of this approach is associated with the need for individual selection of parameters of controlled rotations for each iteration step within the phase estimation algorithm. An alternative to this selection is the use of the Trotter formula [14], which requires a sharp increase in the number of two-qubit gates, and ultimately leads to a decrease in accuracy [3].

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