

Using effective operators in calculating the hyperfine structure of atoms

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We propose a method for calculating the hyperfine structure (hfs) of multielectron atoms based on a combination of configuration superposition and many-body perturbation theory. The latter is used to construct an effective Hamiltonian and an effective hfs operator in configurational space. The method can be applied in calculations of the matrix elements of any one-electron operators. By way of an example we calculate the magnetic hfs constant A for several lowest levels of neutral thallium. We show that the method achieves a calculation accuracy of about 1%, which earlier was possible only for atoms with a single valence electron. © 1998 American Institute of Physics. [S1063-7761(98)00611-8]

1. INTRODUCTION

Recently we proposed a method for calculating the lowest energy levels of multielectron atoms.¹ The calculations done for Tl (Ref. 1) and Ca, Sr, Ba, and Yb (Refs. 2 and 3) demonstrated its effectiveness. In this paper we wish to show that the method can be used to calculate not only energies but also other observables, such as the hyperfine structure (hfs) constants and transition amplitudes. For the sake of definiteness we focus on calculations of the magnetic dipole hfs constant. Generalization to other one-electron operators is obvious. Not that hfs calculations are one of the main tests in calculating amplitudes that do not conserve spatial parity.

At present several methods for calculating multielectron atoms are available. For atoms with one electron in addition to the electrons in filled shells there is the many-body perturbation theory in the residual Coulomb interaction (see, e.g., Ref. 4). For atoms with several valence electrons there is the configuration-superposition method and the multiconfiguration Hartree–Fock method.⁵ Lately the coupled-cluster method has gained wide acceptance.^{6–8} The hfs constants have been repeatedly calculated by all these methods (see, e.g., Refs. 9–14).

The most complicated problem encountered in atomic calculations is the need to correctly account for the correlations between valence electrons and the correlations incorporating core electrons. Correlations of the first type are too strong to be accounted for by ordinary many-body perturbation theory. However, if the number of the valence electrons is not too large, these correlations are taken into account fairly well by the configuration-superposition method or the multiconfiguration Hartree–Fock method. Correlations of the second type are accounted for more simply by many-body perturbation theory, since the number of configurations needed to describe these correlations by the configuration-superposition method or the multiconfiguration Hartree–Fock method is too large.

All this suggests a combination of the configuration-superposition method and many-body perturbation theory. The latter is used to set up an effective Hamiltonian for the valence electrons. After that the Schrödinger equation with the effective Hamiltonian can be solved by the configuration-superposition method. At this stage only valence electrons are accounted for explicitly. In Ref. 1 we showed that the Brillouin–Wigner perturbation theory and the ordinary diagrammatic technique are sufficient for constructing the effective Hamiltonian. In the present paper we discuss the setting-up of effective Hamiltonians for other observables.

In Sec. 2 we define the valence subspace and give the main formulas for the effective Hamiltonian. Section 3 is devoted to a discussion of other effective operators. In Sec. 4 we calculate the hyperfine structure in thallium.

2. EFFECTIVE HAMILTONIAN FOR VALENCE ELECTRONS

Here we are interested in low-energy atomic states with energies $E_i - E_0 < \varepsilon$, where E_0 is the ground-state energy of the atom. Then to a first approximation we can assume that the inner electrons, whose Hartree–Fock energies ε_n are much higher (in absolute value) than ε , form a core, which is described by the wave function

$$\Psi_{\text{core}} = (N_c!)^{-1/2} \det(\phi_1, \phi_2, \dots, \phi_{N_c}), \quad (1)$$

$$h_{\text{HFD}} = \varepsilon_n \phi_n, \quad (2)$$

where h_{HFD} is the Hartree–Fock–Dirac operator. Although this operator is used to define the atomic core, it may incorporate the field of all the valence electrons or of several such electrons. For instance, below we examine thallium as an atom with three valence electrons and the core $[1s^2 \dots 5d^{10}]$, while the h_{HFD} operator is set up for the $1s^2 \dots 5d^{10}6s^2$ configuration (the V^{N-1} approximation, where N is the number of electrons in the atom).

We call the subspace of the multielectron states Ψ for which the core electrons are in state (1) the valence subspace and introduce the projector P on this subspace. The complementary subspace, with a corresponding projector $Q = 1 - P$, is characterized by the fact that at least one of the core electrons is excited to states lying higher than ϕ_{N_c} .

The Schrödinger equation

$$H\Psi = E\Psi \quad (3)$$

can be shown¹ to be equivalent to the following equation in the P space for the function $\Phi = P\Psi$:

$$[PHP + \Sigma(E)]\Phi = E\Phi, \quad (4)$$

$$\Sigma(E) = PV'R_Q(E)V'P, \quad (5)$$

where V' is the operator of the residual Coulomb interaction, and $R_Q(E)$ is the Green's function in the Q space, i.e.,

$$V' \equiv H - H_0, \quad (6)$$

$$R_Q(E) = Q \frac{1}{E - QH_0Q} Q. \quad (7)$$

The operator H_0 can be expressed in terms of the operator in Eq. (2) as follows:

$$H_0 = \sum_{i=1}^N h_{\text{HFD}}(\mathbf{r}_i) - W. \quad (8)$$

The constant W appears on the right-hand side of this equation because the sum of single-particle energies ε_n allows for the electrostatic interaction of the electrons between each other twice and cannot serve as a good approximation for the total energy E of the atom. This constant can be found from the condition¹

$$\langle \Psi_{\text{core}} | H_0 | \Psi_{\text{core}} \rangle = \langle \Psi_{\text{core}} | H | \Psi_{\text{core}} \rangle \equiv E_{\text{core}}, \quad (9)$$

or $\langle \Phi_0 | H_0 | \Phi_0 \rangle = \langle \Phi_0 | H | \Phi_0 \rangle$, where Φ_0 is the ground-state wave function of the atom. Finally, this constant can be used as an adjustable parameter, selected by the best match between theory and experiment. Note that in the lowest perturbation order for the operator $\Sigma(E)$, the redefinition of this constant as $W \rightarrow W + \delta$ is equivalent to a shift in energy: $\Sigma(E) \rightarrow \Sigma(E + \delta)$.

The solutions of Eqs. (3) and (4) are related by

$$\Psi = [P + R_Q(E)V'P]\Phi, \quad (10)$$

which implies $\Phi = P\Psi$.

The orthonormalization condition $\langle \Psi_i | \Psi_k \rangle = \delta_{i,k}$ can be approximately reduced to a condition imposed on the functions Φ ,

$$\langle \Phi_i | 1 - \partial_E \Sigma(\bar{E}) | \Phi_k \rangle \approx \delta_{i,k}, \quad (11)$$

where $\bar{E} \approx (E_i + E_k)/2$. Note that of the equations in (4)–(11) only the last is an approximation.

It is natural to call the operator in the square brackets in Eq. (4) the effective Hamiltonian H_{eff} for the valence electrons. Equations (4)–(7) make it possible to use this Hamiltonian for implementing the usual methods of many-body perturbation theory and, in particular, the diagrammatic

technique.^{1,2} To this end we use the representation of the exact Green's function in terms of the Green's function in the Hartree–Fock–Dirac representation:

$$R_Q(E) = R_Q^0(E) + R_Q^0(E)V'R_Q(E), \quad (12)$$

$$R_Q^0(E) = Q \frac{1}{E - QH_0Q} Q. \quad (13)$$

3. EFFECTIVE OPERATORS FOR VALENCE ELECTRONS

We assume that we know the solutions of Eq. (4), which we will use to find an observable a corresponding to the one-electron operator A :

$$a = \langle \Psi | A | \Psi \rangle. \quad (14)$$

We define an effective operator A_{eff} such that

$$a = \langle \Phi | A_{\text{eff}} | \Phi \rangle. \quad (15)$$

Combining (10), (14), and (15), we arrive at an expression for A_{eff} :

$$A_{\text{eff}} = PAP + PV'R_Q(E)AP + PAR_Q(E)V'P + PV'R_Q(E)AR_Q(E)V'P. \quad (16)$$

Equations (12) and (16) allow the operator A_{eff} to be expanded in a power series in V' . Unfortunately, in most cases this series converges very slowly, so that usually instead of the consistent perturbation-theory approach one uses approximations that partially allow for all orders in V' . The random-phase approximation (RPA) is the one most often used in this case (see, e.g., Ref. 15). Let us see how this approximation agrees with Eq. (16).

On the right-hand side of Eq. (16), the operator A is always combined with the exact Green's function. We introduce a new operator \tilde{A} such that

$$\begin{pmatrix} A_{PP} & A_{PQ}R_Q \\ R_Q A_{QP} & R_Q A_{QQ} R_Q \end{pmatrix} = \begin{pmatrix} \tilde{A}_{PP} & \tilde{A}_{PQ} R_Q^0 \\ R_Q^0 \tilde{A}_{QP} & R_Q^0 \tilde{A}_{QQ} R_Q^0 \end{pmatrix}, \quad (17)$$

where $A_{PP} \equiv PAP$, etc. If we were able to construct \tilde{A} , we would easily derive the operator A_{eff} , since by substituting \tilde{A} into Eq. (16) we arrive at a situation in which all exact Green's functions are replaced by Hartree–Fock Green's functions. Equation (17) is equivalent to the following system of operator equations:

$$\tilde{A}_{PP} = A_{PP}, \quad (18)$$

$$\tilde{A}_{PQ} = A_{PQ} + \tilde{A}_{PQ} R_Q^0 V' Q, \quad \tilde{A}_{QP} = \tilde{A}_{QP}^\dagger, \quad (19)$$

$$\begin{aligned} \tilde{A}_{QQ} = & A_{QQ} + \tilde{A}_{QQ} R_Q^0 V' Q + Q V' R_Q^0 \tilde{A}_{QQ} \\ & - Q V' R_Q^0 \tilde{A}_{QQ} R_Q^0 V' Q. \end{aligned} \quad (20)$$

The RPA equations for the core electrons (Fig. 1) resemble Eqs. (19) and (20). The main difference between the operator \tilde{A} and the RPA operator A_{RPA} is that the former is not single-particle. Moreover, the random-phase approximation does not incorporate a number of single-particle corrections allowed by Eqs. (19) and (20). However, the most important terms of Eqs. (19) and (20) are taken into account by A_{RPA} .

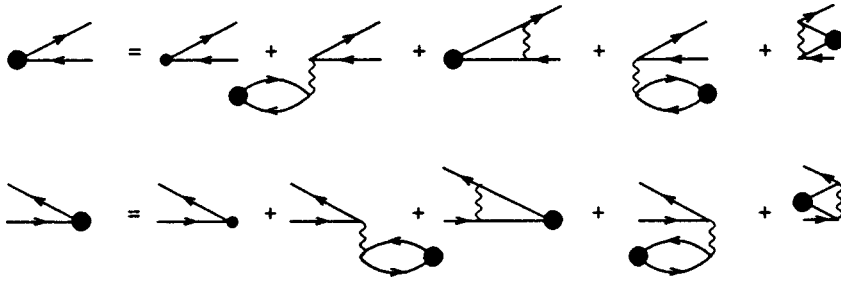


FIG. 1. Diagrammatic representation of the random-phase approximation method for the core. A small filled circle stands for the bare matrix element of the one-electron operator A , and a large filled circle corresponds to a similar matrix element in the random-phase approximation. The wavy line stands for the Coulomb interaction.

All this implies that we can write an approximate equation for the effective operator (16),

$$A_{\text{eff}} \approx PAP + PV'R_Q^0(E)A_{\text{RPA}}P + PA_{\text{RPA}}R_Q^0(E)V'P. \quad (21)$$

The equation does not contain a term similar to the fourth term on the right-hand side of Eq. (16). This term is partially included in the second and third terms. The remaining part corresponds to what is known as structural radiation, which in most cases is very weak.¹⁴

Equation (21) also resembles the RPA equation for valence electrons. However, in addition to the RPA diagrams (Fig. 2a), this equation contains two additional types of diagram (Figs. 2b and 2c). The diagrams of Fig. 2b can be called subtractational (SBT) by analogy with similar diagrams for the operator Σ (Ref. 1). They appear only when the Hartree-Fock operator, which incorporates the field of (several) valence electrons, is used to solve the RPA equations. It should be recalled that such an operator is used in setting up the core wave function (1). The diagrams in Fig. 2c correspond to the two-particle corrections A_{TP} to the effective operator. There is also an important type of diagram (Fig. 2d) not included in the approximation (21). Such diagrams refer to a higher order in the many-body perturbation theory,

but their contribution is extremely large, while similar diagrams in which the A_{RPA} vertex is connected to the other part of the diagram by a particle line rather than by a hole line are taken into account when (21) is substituted in (15).

Combining all these corrections, we arrive at an approximate expression for the effective operator:

$$A_{\text{eff}} \approx P(A_{\text{RPA}} + A_{\text{SBT}} + A_{\text{TP}} + A_{\sigma})P, \quad (22)$$

where the four terms on the right-hand side correspond to the four types of diagram in Figs. 2a–2d. Below, in calculating the hfs constants, we use Eqs. (15) and (22). We note once more that this approximation allows only for the first order of many-body perturbation theory. Even in second order there are corrections of the structural-radiation type, which are not included in (22). On the other hand, Eq. (22) takes into account some of the most important higher-order corrections.

To conclude this section we note that the operator A_{eff} is used to solve Eq. (4). This means that excitations of valence electrons are taken into account in all orders, which guarantees an accuracy higher than that achieved by many-body perturbation theory.

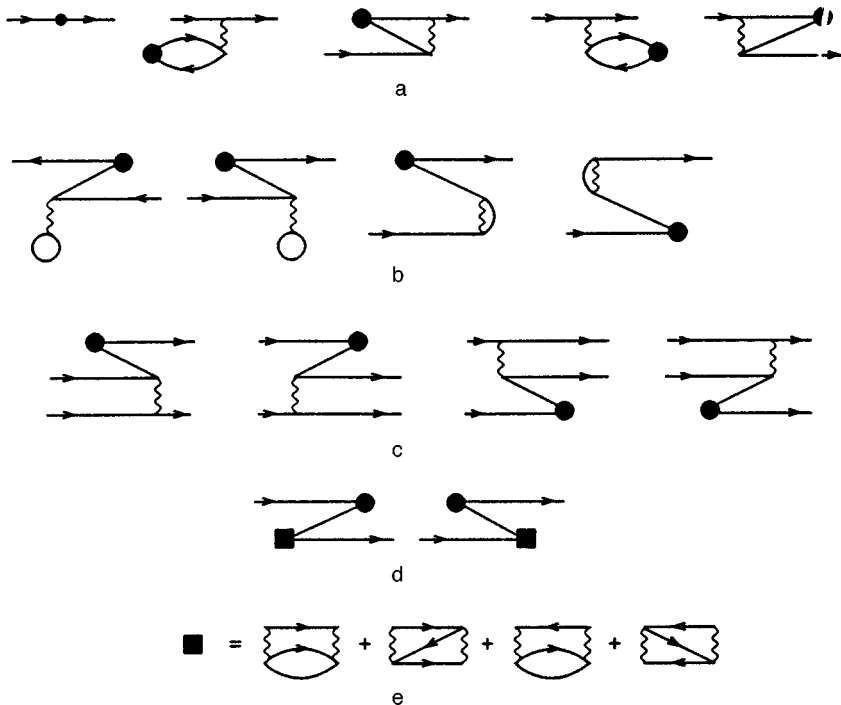


FIG. 2. Diagrammatic representation of the effective operator A_{eff} for the valence electrons: (a) the random-phase approximation A_{RPA} , (b) the subtractational correction A_{SBT} , (c) the two-particle correction A_{TP} , and (d) the self-energy correction A_{σ} . A filled square stands for the self-energy block (e).

TABLE I. Valence energies of several lowest levels of Tl.

Level	E_{val} , a.u.	ΔE , cm ⁻¹	
		Calculation	Experiment ¹⁸
$6p_{1/2}$	-2.072 084	0	0
$6p_{3/2}$	-2.036 471	7808	7793
$7s_{1/2}$	-1.951 435	26 472	26 478
$7p_{1/2}$	-1.916 363	34 169	34 160
$7p_{3/2}$	-1.911 804	35 170	35 161
$6d_{3/2}$	-1.907 348	36 148	36 118
$6d_{5/2}$	-1.907 011	36 222	36 200

Note: The effective Hamiltonian was set up for the energy $E_{\text{val}} = -1.64$ a.u., which was chosen on the grounds of best agreement between that the calculated energy intervals between the levels and the experimental values of these intervals.

4. CALCULATIONS OF THE HYPERFINE STRUCTURE

In the point-nucleus approximation, the operator of the magnetic hyperfine structure has the form

$$H_{\text{hfs}} = \frac{\mu_n \mu_0}{I} \frac{\mathbf{I} \cdot (\boldsymbol{\alpha} \times \mathbf{r})}{r^3}, \quad (23)$$

where μ_n is the magnetic moment of the nucleus, μ_0 is the Bohr magneton, and $\boldsymbol{\alpha}$ is the Dirac matrix. For a finite nucleus, this expression can be used only if $r > r_n$ holds, where r_n is the radius of the nucleus. Inside the nucleus the hfs operator depends on the nuclear structure rather than on the magnetic moment of the nucleus. The exact formulas can be found in Ref. 13. There it is also shown that the corresponding corrections are extremely small, so that here we use an approximation in which inside the nucleus the dependence of the operator (23) on the radius, $1/r^3$, is replaced by r/r_n^4 . Such an approximation is quite sufficient if we need an accuracy of about 1%.

We begin our calculations of the hyperfine structure of the thallium atom by constructing an effective Hamiltonian and solving Eq. (4). This part of our calculations differs somewhat from the procedure adopted in Ref. 1. We enlarged the basis set of the radial functions, which now includes orbitals up to $21s$, $21p$, $21d$, $18f$, $18g$, and $14h$ (the procedure of constructing orbitals is similar to the one described in Refs. 1, 16, and 17). We also significantly increased the number of configurations. Moreover, in calculating the diagrams for the operator $\Sigma(E)$ we also calculated their first energy derivatives. This made it possible to allow in the first approximation for the energy dependence of the matrix elements of the effective Hamiltonian. The thallium spectrum calculations employed an effective Hamiltonian for different valence electrons, which, as noted earlier, corresponds to different constants W in (8). We found that the agreement with the experimental spectrum is best when $\Sigma(E)$ is taken at an energy $E_{\text{val}} = E - E_{\text{core}} = -1.64$ a.u. (see Table I).¹⁾ What is important is that the low-energy part of the spectrum of neutral thallium was well reproduced by these calculations and the valence energy of the ground state was found to coincide perfectly with the experimental value of 2.0722 a.u.¹⁹⁾

The next step involved constructing the effective operator (22). All corrections except A_{TP} are determined by single-particle operators and can be reduced to calculating effective one-electron integrals. Calculating the expectation value (15) for such operators is easy. However, allowing for the two-electron correction A_{TP} required more involved calculations. Hence in calculating these corrections we discarded the contributions of configurations whose weight in the wave function was no larger than 10^{-4} .

To determine the error associated with the incompleteness of the configuration-superposition method, we estimated the contribution of virtual orbitals not included in the superposition of configurations in the lowest perturbation order. We call the corresponding corrections the valence corrections. These corrections are important only for the $6p_{3/2}$ and $7p_{3/2}$ levels, where the other corrections are small or cancel each other almost perfectly.

Finally, the last correction arises if we allow for the conditions (11). For the lowest levels with fixed angular momentum J and parity P the conditions simply determine the corrections to normalization. For excited states, strictly speaking, we must allow for violation of orthogonality with the lowest states, but here we took into account only the normalization condition. The results of calculating the hyperfine structure of thallium are listed in Table II.

5. CONCLUSION

A comparison of the results listed in Table I and those of Ref. 1 shows that even approximate allowance for the energy dependence of the operator Σ combined with an optimum choice of the constant W , which determines the starting approximation (8) of the Hamiltonian, make it possible to enhance the accuracy of calculations of the atomic spectrum. The largest deviation from the experimental data in such calculations amounts only to 0.2%, while in the previous calculations it amounted to about 1%. This refinement of the spectrum was found to have an effect on the results of hfs calculations.

The results listed in Table II show that the Hartree–Fock–Dirac approximation for the magnetic hfs constant agrees very poorly with the experiment. In some cases it even yields an incorrect sign or an incorrect order of magnitude of the constant. Allowing only for configuration superposition does not improve the results significantly.

Only when many-body perturbation theory and the configuration-superposition method are combined does the accuracy improve substantially. Many-body perturbation theory yields three types of correction, resulting from (i) replacing the valence Hamiltonian by the effective Hamiltonian (allowance for $\Sigma(E)$), (ii) using the effective hfs operator (A_{SBT} , A_{TP} , and A_{σ}), and (iii) the normalization condition (11). The last correction is less than 1%, while the other two are much larger.

Note that the various many-body perturbation-theory corrections often cancel one another because summation over intermediate states leads to contributions that violate the Pauli exclusion principle. Such contributions, obviously, cancel each other, so that their net effect is zero. For in-

TABLE II. Magnetic hfs constants for several lowest levels of ^{205}Tl (MHz).

	$6p_{1/2}$	$6p_{3/2}$	$7s_{1/2}$	$7p_{1/2}$	$7p_{3/2}$	$6d_{3/2}$	$6d_{5/2}$
HFD	17 554	1302	7612	1957	188	21	9
CS	+ 195	- 1369	+ 3655	- 88	+ 114	- 149	+ 307
$\Sigma(E)$	+ 3197	+ 48	+ 421	+ 290	- 41	+ 90	- 135
RPA	+ 1359	+ 327	+ 1043	+ 134	+ 71	+ 6	+ 14
A_{SBT}	- 1225	- 120	- 72	- 116	- 17	- 16	- 11
A_{TP}	+ 1130	+ 44	- 10	+ 105	+ 6	+ 12	+ 8
A_{σ}	- 1090	- 19	- 303	- 105	- 10	+ 2	- 6
Valence	- 13	+ 53	+ 43	- 6	+ 6	- 1	- 1
Normalization	- 214	- 2	- 82	- 14	- 2	0	- 1
Theory of Ref. 14	21 760	- 1919	12 470	2070	195		
Theory of Ref. 13	21 300	339	12 760				
Present work	21 623	264	12 307	2157	315	- 35	184
Experiment	21 311	265	12 297	2155	309	- 43	229

Note: We list the values of the hfs constants in the Hartree–Fock–Dirac (HFD) approximation and the correction yielded by configuration superposition (CS) and the correction Σ to the Hamiltonian. We allow for the corrections to the hfs operator arising from the use of the random-phase approximation (RPA), the subtractional correction A_{SBT} , the two-particle correction A_{TP} , the self-energy correction A_{σ} , the valence correction reflecting the incompleteness of configuration substitution, and the normalization correction.

stance, the sum $A_{\text{RPA}} + A_{\text{SBT}} + A_{\text{TP}}$ obeys the Pauli exclusion principle, while each term in it does not. For this reason the separate contributions to this sum have no physical meaning. Moreover, in most cases there is partial balance between the correction due to the use of an effective Hamiltonian and the correction A_{σ} . In our case this balance results from the fact that the contributions of the intermediate electron and hole state have different signs but comparable values. As a result, the net many-body perturbation-theory correction is much smaller than the separate contributions.

The data in Table II suggest that for all the levels except the d levels the agreement with experiment is extremely good. The largest many-body perturbation-theory corrections arise for the $6p_{1/2}$ level, where they amount to more than 3000 MHz. This explains the lower accuracy achieved in calculating this constant (1.4%). Two other large constants (for the $7s$ and $7p_{1/2}$ levels) were found to be calculated with an accuracy ten times higher, which corresponds to appreciably smaller values of the many-body perturbation-theory corrections. For the $6p_{3/2}$, $6d_{3/2}$, and $6d_{5/2}$ levels we have an extremely poor starting approximation. Hence the perfect agreement between our results for the $6p_{3/2}$ level and the experimental data can to a certain extent be considered a coincidence. Note that to obtain a correct result we needed to allow for all the corrections here.

On the whole we can say that the adopted method proved to be very effective in calculating the spectrum and the hfs constants. Note that in a somewhat simplified form this method has recently been used to calculate the hfs constants and P, T -odd matrix elements for the BaF molecule,²⁰ with the accuracy of this method also increasing significantly. Further refinements require increasing the number of higher-order corrections taken into account. This can be done by resorting to the methods used earlier in calculations for the cesium and thallium atoms by a pure perturbation-theory approach.^{14,21} First one must allow for higher-order correc-

tions to the operator Σ . The leading corrections of this type correspond to the inclusion of a polarization operator in phonon lines. To first order these corrections can be taken into account by introducing screening coefficients (see, e.g., Ref. 3). One must also allow for the correction to the effective hfs operator related to structural radiation.

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¹The problem of selecting the energy is discussed in greater detail in a paper submitted to the journal *Optika i Spektroskopiya* (translated as *Optics and Spectroscopy*).

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