[15]T. A. Carlson, Phys. Rev. 156, 142 (1967).
[16]J. A. R. Samson and G. N. Haddad, Phys. Rev. Lett. 33, 875 (1974).
[17]V. V. Afrosimov, M. Ya. Amusia, Yu. S. Gordeev, M. P. Kazachkov, and S. G. Shchemelinin, Abstracts of Papers VIII ICPEAC 1, Belgrade, 1973, p. 415.
[18]V. V. Afrosimov, I. P. Gladkovskiĭ, Yu. S. Gordeev, I. F. Kalinkevich, and N. V. Federenko, Zh. Tekh. Fiz. 30, 1456 (1960) [Sov. Phys. Tech. Phys. 5, 1378 (1961)].
[19]A. A. Petushkov and A. S. Melioranskiĭ, Peredovoĭ nauchno-tekhnicheskiĭ i proizvodstvennyĭ opyt (Progressive Technical and Manufacturing Practice), No. 5, Leningrad, 1960, p. 18.
[20]S. G. Shchemelinin and V. V. Bagaev, Prib. Tekh. Eksp. No. 2, 247 (1975).
[21]S. G. Shchemelinin and E. P. Andreev, Zh. Tekh. Fiz. 45, 1490 (1975) [Sov. Phys. Tech. Phys. 20, 941 (1975)].
[22]V. V. Afrosimov, Yu. S. Gordeev, M. H. Panov, and N. V. Fedorenko, Zh. Tekh. Fiz. 34, 1613 (1964) [Sov. Phys. Tech. Phys. 9, 1248 (1965)]; V. V. Afrosimov, Yu. A. Mamaev, M. N. Panov, V. Uroshevich, and N. V. Fedorenko, Zh. Tekh. Fiz. 37, 550 (1967) [Sov. Phys. Tech. Phys. 12, 394 (1967)].
[23]M. Ya. Amusia and N. A. Cherepkov, Case Studies in Atomic Physics 5, 47 (1975).
[24]L. V. Chernysheva, M. Ya. Amus'ya, and S. I. Sheftel', Sistema matematicheskogo obespecheniya atomnykh raschetov "ATOM" III. Programma vychisleniya OCO atomov s volnovymi funktsiyami Khartri–Foka i s uchetom mnogoelektronnykh korrelyatsiĭ v odnom perekhode (The "ATOM" III Atomic Computation System Program for the Calculation of Generalized Oscillator Strengths with Hartree–Fock Wave Functions and with Allowance for Many-Electron Correlations in One Transition), Preprint FTI-493, Leningrad, 1975.
[25]L. V. Chernysheva, M. Ya. Amys'ya, and S. I. Sheftel', Sistema matematicheskogo obespecheniya atomnykh raschetov "ATOM" IV. Programma vychisleniya OCO atomov s uchetom mnogoelektronnykh korrelyatsiĭ v dvukh perekhodakh (The "ATOM" IV Atomic Computation Method Program for Calculating the Generalized Oscillator Strengths with Allowance for Many-Electron Correlations in Two Transitions), Preprint FTI-495, Leningrad, 1975.
[26]E. Weigold, S. T. Hood, and P. J. O. Teubner, Phys. Rev. Lett. 30, 475 (1973).
[27]J. A. R. Samson, Adv. in Atomic and Molecular Physics 2, 177 (1966).
[28]Y. B. West, G. V. Marr, Y. Hamley, and K. Codling, Fourth Intern. Conf. on Vacuum-Ultraviolet Radiation Physics, Hamburg, 1974, p. 176.

Translated by J. G. Adashko

Three-photon ionization of metastable helium atoms

J. Bakos,[1] N. B. Delone, A. Kiss,[1] N. L. Manakov,[2] and M. L. Nagaeva

P. N. Lebedev Physics Institute, USSR Academy of Sciences, Moscow

(Submitted March 3, 1976)
Zh. Eksp. Teor. Fiz. 71, 511–525 (August 1976)

Direct and resonance three-photon ionization processes are investigated for helium atoms in 2^1S and 2^3S metastable states. The absolute ionization cross sections, the cross section ratios for plane and circularly polarized radiation at given frequency, and the ratios of ionization cross sections from the 2^1S and 2^3S states have been measured for the direct process. All the measured quantities are in good agreement with calculations performed with the aid of time-dependent perturbation theory. A change in the energy of the resonance transition, which is proportional to the radiaion intensity, has been found for the ionization process in the presence of the two-photon intermediate 2^1S–6^1S resonance. The origin of perturbations of resonance states is discussed.

PACS numbers: 32.10.Vc, 32.10.Qy

1. INTRODUCTION

The ionization of atoms by high-intensity light has recently attracted the attention of both experimenters and theoreticians. Most of the published investigations have been concerned with the ionization of atoms from the ground state.[1] The ionization of atoms from excited states is, however, also of considerable interest. In some cases, it enables us to investigate phenomena which cannot be observed in the case of ionization of atoms from the ground state.

Multiphoton transitions in the spectra of noble gases are an example of this. The high ionization energy of these atoms in the ground state ensures that ionization by optical radiation is highly nonlinear ($k = 10–20$), so that the field strength must be high and the adiabatic parameter low ($\gamma \approx 1$[1]). Under these conditions, the spectrum of excited states is substantially perturbed by the field, so that the systematics of levels of the undisturbed atoms is modified and this, among other things, complicates the interpretation of resonances that appear during the multiphoton ionization process.

On the other hand, because of the large energy gap between the ground and excited states in the spectra of noble gases, ionization from the first excited state is much less nonlinear ($k = 3–5$). The necessary field strengths are therefore relatively low, and one can investigate in the light field the perturbations of atomic states characterized by the same quantum numbers as in the absence of the field. Since, in this case, the parameter γ is much greater than unity, it is reasonable to expect that perturbation theory should be valid for the theoretical description of ionization from excited states.

In this paper, we shall investigate three-photon ion-

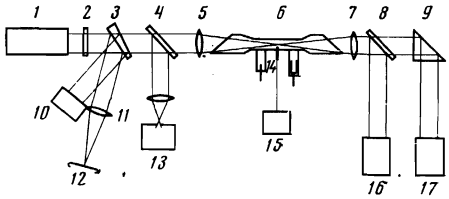

FIG. 1. Schematic diagram of the apparatus: 1) laser; 2) quarter-wave plate; 3), 4), 8) beam-splitting wedges; 5), 7), 11) lenses; 6) discharge tube filled with helium; 9) rotating prism; 10) coaxial photocell; 12) photographic plate; 13) calorimeter; 14) probe; 15) differential amplifier; 16) spectrograph; 17) Fabry-Perot interferometer.

ization of helium atoms from the singlet and triplet metastable states. We have used plane and circularly polarized radiation from a tunable ruby laser.

The ionization energies of helium atoms in metastable states are $I(2^1S) = 3.98$ eV and $I(2^3S) = 4.77$ eV. Using a comparison between the spectra of bound electronic states of the helium atom and the energy of the radiation emitted by the ruby laser ($\hbar\omega \approx 1.78$ eV), and the selection rules for multiphoton transitions, one can show that ionization from the 2^3S state by plane and circularly-polarized radiation, and from the 2^1S state by circularly polarized radiation, is a direct process and no intermediate resonances are produced. One would hope to be able to observe the intermediate two-photon resonance with the 6^1S state by varying the ruby-laser frequency during ionization from the 2^1S state by plane-polarized radiation. Thus, the observation of the above three-photon process can provide experimental data both on the direct and the resonance ionization processes, and, in the second case, on the perturbation of the atomic spectrum in the light field.

2. FORMULATION OF EXPERIMENT

The formulation of the experiment was fairly standard in its general features[1] (Fig. 1). The atoms under investigation were illuminated by the laser radiation in metastable states. The number of resulting ions was then observed as a function of the intensity, frequency, and degree of polarization of the illuminating radiation. The specific feature of this particular experiment was the use of the helium plasma in the afterglow of the discharge as the target, and the utilization of a single probe for measuring the variation in the degree of ionization of the plasma under the influence of the laser radiation.

1. Target

The main requirement that has to be satisfied by the target is that it should contain the maximum density of atoms in metastable states. We have used the optimum conditions from this point of view, i.e., helium-gas pressure of about 1 Torr and discharge-tube diameter of about 1 cm. A constant current of about 3 mA was passed through the tube and could be cut off in a time of about 1 μsec. During the time taken to turn off the discharge, all the highly excited states in the plasma are found to decay, and only ions, electrons, and helium atoms in the ground and excited states remain, together with the helium molecules in the metastable states.

a. Helium atoms in metastable states. The density of atoms in metastable states was measured in the discharge by the standard method involving the determination of the absorption coefficient for light from an auxiliary source, the discharge in which was completely identical to that in the discharge under investigation.[2] Measurements showed that the atom densities averaged over the cross section of the discharge tube were $n(2^1S) = 10^{10.5\pm0.5}$ cm^{-3} and $n(2^3S) = 10^{11.0\pm0.5}$ cm^{-3}.

The lifetimes of metastable atoms in the afterglow plasma were determined by measuring the dependence of the absorption coefficient for light of resonance wavelength ($\lambda_1 = 3889$ Å and $\lambda_2 = 5016$ Å) on time after the discharge was turned off. It was found that $\tau(2^1S) = (39.3 \pm 3.3)$ μsec and $\tau(2^3S) = (81.5 \pm 8.8)$ μsec. These densities and lifetimes of metastable atoms are typical for the above discharge-tube geometry and discharge current.

b. Electrons. The electron density in plasma was determined from the cutoff point on the current-voltage characteristic of a single probe. It was found that $n_e \approx 10^{11}$ cm^{-3}. The electron temperature rises to room value during the discharge switching time.

c. Helium molecules in the metastable state. The data of other experiments (see, for example,[3,4]) suggest that, under our conditions, the density of molecules in metastable states is of the same order as the density of atoms in the 2^1S and 6^1S states. Extrapolation of experimental data reported by Phelps[3] to the parameter values corresponding to our plasma shows that the lower limit for the lifetime is $\tau(He_2^*) = 200$ μsec, which substantially exceeds the lifetime of atoms in metastable states, and the difference lies well outside experimental error.

2. Laser radiation

We used a ruby laser with Q-switching produced by a rotating prism. A large number of longitudinal and transverse modes was generated under the working conditions. The generation frequency could be varied continuously in the range 14 402–14 425 cm^{-1} by varying the temperature of the ruby. Absolute measurements of the laser frequency were carried out with the aid of a diffraction-grating spectrograph. The dispersion of the spectrograph in the region of the ruby-laser radiation was about 1.5 Å/mm. The laser linewidth was measured with a Fabry–Perot interferometer having a resolution of about 0.1 cm^{-1}, and this showed that the laser line was, in fact, a partially resolved doublet with a width of about 0.2 cm^{-1}.

The laser radiation was 99% plane polarized. A quarter-wave quartz plate was placed in the laser beam just in front of the discharge tube. Different degrees of elliptical polarization could be produced by rotating this plate about the beam axis. This method of measuring the degree of ellipticity did not affect the distribution in space of the beam intensity. The degree of

ellipticity of the laser radiation was controlled by a Glan prism, using the readings of a calorimeter.

The absolute intensity of the laser radiation in the region in which the ions were produced was determined by the standard method[11] involving the determination of the energy per laser pulse transmitted by the discharge tube, the distribution in space of the illuminance, and the time distribution of the intensity integrated over the cross section of the beam. Since the laser generated a large number of modes, special measurements were carried out to determine the time distribution of intensity at different points in the cross section of the beam. These measurements showed that the time distributions were the same to within ± 10% at all points in the beam. A lens with a focal length of 400 mm was used to focus the radiation. The transverse size of the laser beam remained practically constant along the axis of the discharge tube. The diameter of the focal spot was about 0.5 mm.

3. The probe method of measurement

Ions produced in the afterglow plasma by the incident laser radiation brought about a change in the probe potential of about 10^{-3} of the plasma potential. Special electronics was developed to balance out the plasma potential and thus isolate the useful signal. The precision of this compensation determined the minimum sensitivity of the probe method, which was 10^8 ions formed in the plasma. The electronics was sufficient to ensure that the useful signal could be measured in a dynamic range of about 10^3. The change U in the probe potential is related to the number N_i of ions produced by the laser radiation by the formula $U = c\xi N_i$, where ξ is the probe efficiency which depends on the initial spatial distribution of the ions generated by the radiation and the distance between the probe and the region in which the ions appear. The absolute probe efficiency can be determined to within ± 50%.

4. The nature of the probe signal

Analysis of the interaction processes between the laser radiation and the afterglow plasma has shown that, for fields $\mathscr{E} < 10^6$ V · cm^{-1}, three-photon ionization of helium atoms and molecules in metastable states can be the source of ions. To verify the validity of this conclusion, we first carried out experiments in which, as the field $\mathscr{E}$ was varied between 2×10^5 and 5×10^5

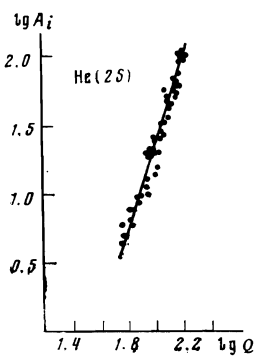

FIG. 2. Probe signal amplitude A_i as a function of energy Q per radiation pulse (logarithmic scales).

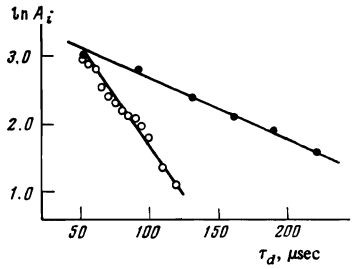

FIG. 3. Typical results of measurements of the probe-signal amplitude A_i as a function of the delay τ_d of the laser pulse relative to the quenched pulse at fixed frequencies: ● —ω = 14 407 cm^{-1}; ○—ω = 14 418 cm^{-1}.

V · cm^{-1}, we determined the probe signal amplitude A_i as a function of the energy Q per pulse of the laser radiation. With constant area S of the focal spot, and constant laser pulse length τ, this dependence is equivalent to the dependence of the ionization probability on the light intensity, $W(F)$. The experiments were performed at frequencies in the range ω = 14 402–14 416 cm^{-1}, where, if the above assumption is correct, the ionization of atoms should be a direct process. Figure 2 shows a typical result of an experiment of this kind. If the relationship between the measured quantities is represented by a power law, the result turns out to be $k = \partial \log A/\partial \log Q = \partial \log W/\partial \log F = 2.9 \pm 0.1$. This clearly confirms the three-photon character of the processes responsible for the formation of the ions.

Effects connected with the ionization of metastable atoms and molecules can be separated because the lifetimes of these states are different. By exposing the afterglow plasma to laser radiation at different instants of time after the discharge has been quenched, it is possible to determine the number of ions as a function of time, and to compare the results of such measurements with the time dependence of the number of metastable atoms or molecules. We have measured the amplitude A_i of the probe signal as a function of the delay time τ_d of the laser pulse relative to the discharge-quenching pulse for constant energy per laser pulse at different frequencies ω in the range ω = 14 407–14 420 cm^{-1}. Figure 3 shows the results of typical measurements of this kind. These data have shown that the time constants characterizing the reduction in amplitude of the probe signal are in good agreement with the corresponding lifetimes of helium atoms in the singlet and triplet metastable states. The precision of these measurements enables us to maintain that the contribution due to the ionization of helium molecules to the probe signal is negligible. We have thus found that ionization from the singlet metastable state of the atom predominates at ω = 14 415–14 420 cm^{-1} whereas ionization from the triplet state predominates at frequencies in the range 14 407–14 410 cm^{-1}.

3. RESULTS OF EXPERIMENTS AND DISCUSSION

1. Direct ionization

a. The conditions for the realization of the direct ionization process are as follows[11]:

$$|E_i - E_0 - k\hbar\omega| > \delta E_{0i}(\mathscr{E}),\ \gamma_{0i}(\mathscr{E}), \qquad (1)$$

where $E_{0,i}$ is the energy of the bound electronic states in the atomic spectrum, γ_{0i} is the reduced width for the 0, i transition, and $\delta E_{0,i}(\mathscr{E})$ and $\gamma_{0i}(\mathscr{E})$ represent variations produced by the field. As a matter of fact, (1) is a condition for a large detuning from resonance in the absence of the field and a small perturbation of the spectrum under the action of the field.

To determine the frequency band in which direct ionization takes place, we have measured the functional dependence of the ionization probability W on frequency ω for fixed light intensity F. Figure 4 shows the probe-signal amplitude A_i as a function of frequency ω for plane and circularly-polarized radiation. It is clear that the two functions are quite different. In the case of the circularly-polarized light, the probability is independent of the radiation frequency to within experimental error, whereas, in the case of plane polarization, there is a resonance at $\omega \approx 14\,420$ cm^{-1}, at which the ionization probability rapidly increases. Similar measurements performed for the ionization from the 2^3S state in the case of linear and circular polarization have shown that the probability is independent of frequency throughout the interval in which a detectable ion yield from this state can be established. These experiments have thus shown that ionization from the 2^1S and 2^3S states by linearly and circularly polarized light is, in fact, a direct process of frequencies in the range $\omega = 14\,402$–$14\,416$ cm^{-1}.

b. Calculation of the direct-ionization probability. Experimental data on the direct ionization process can be compared with calculations performed with the aid of time-dependent perturbation theory. This method[5] shows that the probability of three-photon ionization from an S state in the case of plane-polarized radiation is

$$W^l(\omega) = 2\pi\left(\frac{F}{F_0}\omega\right)^3 \left\{\frac{1}{27}\left[T_{10} + \frac{4}{5}T_{12}\right]^2 + \frac{4}{175}T_{32}^2\right\}, \qquad (2)$$

whereas, in the case of circularly-polarized radiation,

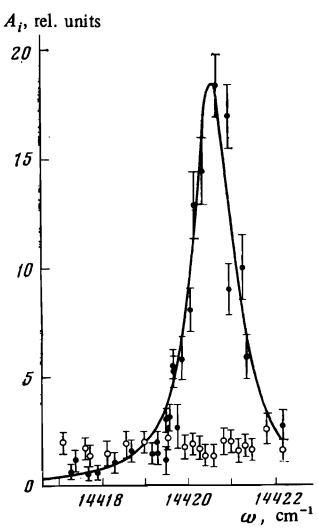

FIG. 4. Probe-signal amplitude A_i as a function of the frequency ω at a constant laser intensity: ●—plane polarization; ○—circular polarization.

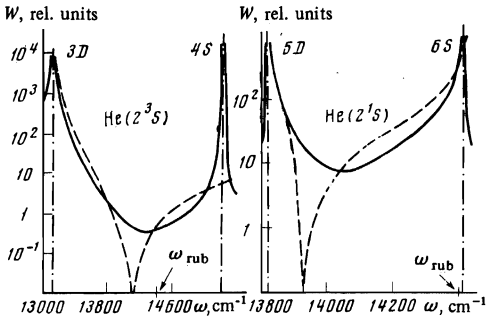

FIG. 5. Frequency dependence of the probability of three-photon ionization of helium atoms from the $2\,^1S$ and $2\,^3S$ states (theoretical calculation from[51]) for plane (solid curve) and circular (broken curve) polarizations: ω_{rub} is the ruby-laser frequency.

$$W^c(\omega) = 2\pi\left(\frac{F}{F_0}\omega\right)^3 \frac{2}{35}T_{32}^2. \qquad (3)$$

In these expressions, F is the flux of photons in cm^{-2} · sec^{-1}, $F_0 = 3.22\times 10^4$ cm^{-2} · sec^{-1} is the atomic unit of flux, $T_{ll'}$ is the radial composite matrix element expressed in terms of the radial Green function $g_l(E; r, r')$ of the optical electron in the atom

$$T_{ll'} = \langle R_l | r_3 g_{l'}(E_i + 2\omega;\ r_3, r_2) r_2 g_{l'}(E_i + \omega;\ r_2, r_1) r_1 | i\rangle,$$

and $|i\rangle$ and $|R_l\rangle$ are the wave functions of the initial and final states of the optical electron.

Numerical calculations of the function $W^{l,c}(\omega)$ are shown in Fig. 5 for one interval between resonances (3^3D–4^3S and 5^1D–6^1S), which contains the ruby-laser frequency. The mean difference between $W(2^1S)$ and $W(2^3S)$ amounts to one or two orders [including the regions near the resonance and frequencies for which $W^c(\omega) \approx 0$], which is to be expected in view of the substantial difference between the structures of the singlet and triplet terms. The substantial increase in $W_l(2^1S)$ and $W^c(2^1S)$, in comparison with $W(2^3S)$ near $\omega \approx 14\,400$ cm^{-1}, is connected with the proximity of the two-photon intermediate resonances 2^1S–6^1S and 2^1S–6^1D for which the detuning is about 30 and about 180 cm^{-1}, respectively.

To calculate $T_{ll'}$, we used the wave functions for the initial 2^1S and 2^3S states, constructed in the quantum-mechanical defect approximation (QMD) and the Hartree–Fock method, whereas the Green function g_l was obtained in the QMD approximation.[5] Comparison of the results of these calculations shows that the choice of the initial-state wave function is important in the case of ionization from the 2^1S state, but plays a minor role in the case of ionization from the 2^3S state. As an example, Table I shows the data obtained for a number of frequencies that are interesting from the point of view of comparison with experimental results.

Comparison between the calculations and experimental data can be carried out, firstly, by comparing the absolute ionization cross sections at constant frequency and polarization; secondly, by comparing the ratio of cross sections at fixed frequency but different polariza-

TABLE I. Data on the direct three-photon ionization process in metastable helium atoms for plane- and circularly-polarized radiation. The dimensions of α are $cm^6 \cdot sec^2$.

	ω, cm^{-1}	Experiment		QMD		HF	
		α_3^l	α_3^l/α_3^c	α_3^l	α_3^l α_3^c	α_3^l	α_3^l/α_3^c
He(2^1S)	14412	10^{-78} $^{+1.6}_{-1.3}$	0.5±0.15	$5.16\cdot10^{-80}$	0.43	$2.94\cdot10^{-78}$	1.04
	14413			$7.06\cdot10^{-80}$	0.57	$5.2\cdot10^{-78}$	1.80
	14414			$1.2\cdot10^{-79}$	0.95	$9.82\cdot10^{-78}$	2.82
	14415			$2.46\cdot10^{-79}$	1.88	$2.0\cdot10^{-77}$	6.5
	14416			$5.86\cdot10^{-79}$	4.34	$4.62\cdot10^{-77}$	14.6
	14417			$1.87\cdot10^{-78}$	13.4	$1.35\cdot10^{-76}$	41.4
He(2^3S)	14407		0.35±0.15	$4.28\cdot10^{-81}$	0.4	$3.42\cdot10^{-81}$	0.49
	14412			$4.34\cdot10^{-81}$	0.46	$5.14\cdot10^{-81}$	0.49

tion; and, thirdly, by comparing the ratio of cross sections for the ionization from the 2^1S and 2^3S states at constant frequency and polarization.

c. Ratio of cross sections for the ionization from singlet and triplet states. The most precise experimental data on the ratio of cross sections for direct three-photon ionization from the singlet and triplet metastable states can be obtained by choosing the radiation frequency so that these cross sections are roughly equal and the ionization process can be observed from both states simultaneously. The experimental data given in Sec. 2.4 indicate the optimum frequency band. Figure 6 shows a typical result of an experiment of this kind. At the frequency $\omega = 14\,412$ cm^{-1} and for laser-pulse delay $\tau_d \geq 100$ μsec, the dominant process is ionization from the 2^3S state whereas, for 50 μsec $\leq \tau_d \leq 100$ μsec, the contributions of ionization from 2^1S and 2^3S states are the same. The ratio of the ion yields is, therefore, determined only by the ratio of the densities of atoms in the metastable states, since the other parameters governing the ionization process (frequency and intensity) are the same. Measurements of the ratios $A_i(2^1S)/A_i(2^3S)$ and $n_0(2^1S)/n_0(2^3S)$ enable us to conclude that, at $\omega = 14\,412$ cm^{-1} and for plane-polarized light, the cross-section ratio is $\alpha_3(2^1S)/\alpha_3(2^3S) = 10 \pm 5.3$. Comparison of this result with the calculations summarized in the table shows that calculations based on the QMD wave functions provide a satisfactory description of the observed ratio, to within experimental error. On the other hand, calculations based on the Hartree–Fock functions predict a value which is greater by a factor of about 1.5 as compared with the measured ratio.

d. Ratio of ionization cross sections for plane- and circularly-polarized light. The ionization probability has been measured for alkali atoms as a function of the polarization of incident radiation in[6,7]. We have determined the ratio of cross sections for three-photon ionization of metastable helium atoms by plane- and circularly-polarized light at frequencies in the range $\omega = 14\,402$–$14\,416$ cm^{-1}. We recall that a change in the degree of polarization does not result in a change in any of the other radiation parameters (Sec. 2.2), so that the ratio A_i^l/A_i^c of probe signals at each fixed radiation frequency is equal to the cross section ratio α_3^l/α_3^c for linear (l) and circular (c) polarizations. We have obtained the following results: $\alpha_3^l/\alpha_3^c = 0.5 \pm 0.15$ (2^1S) for frequencies ω between 14 412 and 14 415 cm^{-1} and $\alpha_3^l/\alpha_3^c = 0.35 \pm 0.15$ (2^3S) at $\omega \approx 14\,407$ cm^{-1}.

If we compare this with the results listed in the table, we see that there is satisfactory agreement between experimental data and calculations based on the QMD wave functions. The Hartree–Fock approximation, on the other hand, leads to a much higher result for the ratio of cross sections for ionization from the 2^1S state. However, in our view, it would be premature to draw any definite conclusions with regard to the inadequacy of the Hartree–Fock wave functions. The absolute probabilities for the 2^1S state are determined by the "resonance" matrix element T_{10}, and the use of the Hartree–Fock wave function for the 2^1S state leads, in this case, to an internal inconsistency in the computational scheme because the intermediate n^1S states which provide the main contribution to T_{10} are Coulomb-like. This "dephazing" of the wave functions may, in fact, be responsible for the above discrepancies. A similar situation occurs, for example, in the calculation of the single-photon matrix element of the form $\langle n_1S|r|n_2S\rangle$ with approximate functions $|n_1S\rangle$ and $|n_2S\rangle$ obtained by different methods. It follows that, to elucidate the accuracy of the QMD approximation in the case of metastable states, we need a situation in which the main contribution to T is provided by virtual states with $l > 0$, which are Coulomb-like with a high degree of accuracy. This case is, in fact, realized in ionization from the triplet state, in which case the results of calculations performed by different methods are in good agreement (see Table I).

e. Measurement of the three-photon ionization cross sections. The measurements necessary to determine the absolute ionization cross section were performed for the case of ionization from the 2^1S state by plane-polarized radiation of frequency $\omega \approx 14\,415$ cm^{-1}. It is known[1] that the cross section α_3 for three-photon ionization is related to the measured parameters by the formula

$$\alpha_3 = \frac{N_i}{n_0}\left(\frac{\hbar\omega}{Q}\right)^3 \frac{S^3\tau^3}{V_3\tau_3}, \tag{4}$$

where N_i is the number of ions produced by the laser radiation, n_0 is the neutral atom density, Q and ω are, respectively, the energy per pulse and radiation frequency,

$$V_3 = \int f^3(x,y,z)\,dv, \quad \tau_3 = \int \psi^3(t)\,dt$$

are the volume of the target and the time of interaction

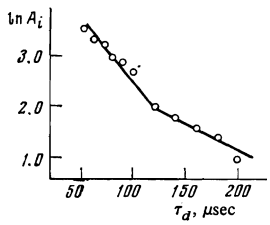

FIG. 6. Probe-signal amplitude A_i as a function of the delay of the laser pulse relative to the quenched pulse at the frequency $\omega = 14\,412$ cm^{-1}.

with the field involving the absorption of three photons in a single event, $f(x, y, z)$ and $\psi(t)$ are the spatial and temporal distributions of the radiation intensity, and S and τ are the area of the focal spot and the length of the laser pulse, respectively.

The measurement errors in the quantities present in (4) are such that only the limits of uncertainty in α_3 can be indicated.[8] For plane-polarized radiation, $\alpha_3 = 10^{-78^{+1.8}_{-1.3}}$ cm^6 · sec^2. Comparison of this result with the results of calculations (see Table I) shows that there is good agreement to within experimental error. It is important to note that results on the ratios of ionization cross sections can be used to obtain absolute ionization cross sections for other polarizations and for the 2^3S state.

Comparison of experimental data on the cross sections for the three-photon ionization of the helium atom from metastable states with calculations (see Table I) indicates that time-dependent perturbation theory can be used to calculate the absolute cross sections with satisfactory accuracy. This conclusion was previously established for the alkali atoms.[6,8] We note that the high precision of the measured cross-section ratios enables us to determine very accurately the ratio of the radial composite matrix elements T and thus determine the accuracy of the various theoretical methods for computing the above quantities which, in multiphoton spectroscopy, play the role of oscillator strengths in linear optics.

2. Resonance ionization

Studies of the resonance multiphoton ionization of atoms enable us to elucidate the physics of the various phenomena that occur during a strong perturbation of the atom by an optical-frequency field. Several workers[9,10] have investigated perturbations of the states of different atoms by plane-, circularly-, and elliptically-polarized radiation. We note that all the results that have been obtained correspond to the case $k_{0r} > 2k_{rE}$, where $0r$ and rE correspond to transitions from the ground state 0 to the excited state r, and from the excited state r into the continuous spectrum E, and k is the degree of nonlinearity of the transition. When this inequality is satisfied, the resonance process has a cascade character.[11] The resonance that can be produced during the ionization of the helium atom from the 2^1S state has a different character, since $k(2^1S-6^1S) = 2k(6^1S-E) = 2$. This process is of great interest because it is not clear, *a priori*, whether the cascade transition or the two-photon resonance mixing of the 2^1S and 6^1S states predominates in this case.[11]

Measurements of the probability $W^I(2^1S)$ as a function of the frequency of the incident radiation (Fig. 4) have shown that, in accordance with the selection rules, the two-photon resonance 2^1S-6^1S takes place, and a sharp increase in the ion yield is observed as the frequency of the incident radiation is varied in the region near resonance. Apart from the very fact that the two-photon resonance does take place, the results of this experiment have also shown that the energy of the 2^1S-6^1S transition undergoes a substantial change in the light field. It is clear from Fig. 4 that the resonance ion yield corresponds to $\omega \approx 14420.7$ cm^{-1} which differs by about 2 cm^{-1} from the resonance frequency $\omega_0 = [E(6^1S) - E(2^1S)]/2\hbar = 14418.7$ cm^{-1} in the absence of the field. This difference is greater by a substantial factor than the half-width of the resonance maximum. Such measurements can therefore provide information on transition energies between bound states of the atom, the width of these states, and the ionization probability as functions of the field, and on the dependence of the probability on the frequency of the incident radiation.

We have carried out measurements of the ionization probability as a function of the incident frequency. These are analogous to the experiments described above (see Sec. 3.1) and were carried out for a number of fixed values of the incident intensity (Fig. 7). To reduce the measurement error, the laser was carefully stabilized, and the spatial and temporal parameters of the incident radiation were held constant for a large number of laser pulses generated in the frequency band between 14419 and 14424 cm^{-1}. By varying the radiation energy with the aid of an absorber mounted in front of the discharge tube, we observed the probe signal amplitude as a function of the radiation frequency for different fixed values of the energy per pulse. This dependence was also measured for different values of the energy, and the spatial and temporal distributions of intensity were monitored. Fluctuations in these distributions were found to affect the spread of the probe signal ($\approx 20\%$) at fixed radiation energy. We note that the frequency corresponding to the maximum in the ion yield (resonance frequency) can be determined to within ± 0.1 cm^{-1}.

a. Change in the energy of the 2^1S-6^1S transition. The dependence of the 2^1S-6^1S transition energy on the incident-light intensity, expressed in relative units, is shown in Fig. 8. The experimental points lie well on the straight line corresponding to the quadratic depen-

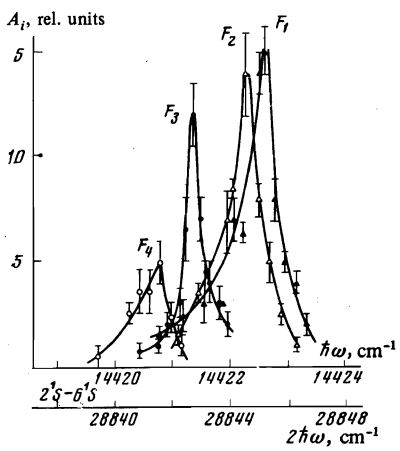

FIG. 7. Measurements of the resonance three-photon ionization of helium atoms from the $2\,^1S$ state. The figure shows the probe-signal amplitude A_i as a function of frequency for four different intensities of plane-polarized light. The lower horizontal scale shows the $2\,^1S$ transition energy in the unperturbed spectrum of the helium atom ($F_1 : F_2 : F_3 : F_4 = 1 : 0.9 : 0.75 : 0.5$).

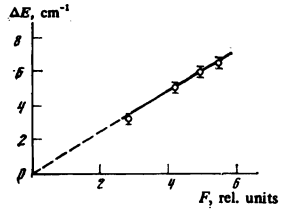

FIG. 8. Change ΔE of the $2\,^1S$–$6\,^1S$ transition energy as a function of the radiation intensity F.

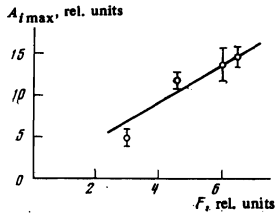

FIG. 9. Probe-signal amplitude at resonance $A_{f\,max}$ as a function of the radiation intensity F.

dence of the change in the transition energy on the field strength: $\Delta E(6^1S-2^1S)=\alpha\mathscr{E}^2$. To determine the absolute magnitude of the coefficient α, we must know the effective value of the radiation intensity that corresponds to the observed maximum ion yield. The effective intensity has to be introduced because the spatial distribution of radiation in the region in which the ions are formed is inhomogeneous, the ionization process is nonlinear, and the integrated ion yield is recorded. It is clear that both large volumes of the target in which the intensity is low, and small volumes of the target in which the intensity is a maximum, provide a small contribution to the integrated ion yield which is largely due to the volume into which a certain effective intensity is introduced, and this was determined in our experiment from experimental data on the spatial distribution of the radiation throughout the target. Numerical calculations on a computer have shown that $F_{eff}=0.55F_{max}$. The value of α for the 2^1S–6^1S transition, determined from experimental data on ΔE for the corresponding $F_{eff}=6.6\times10^{26}$ cm$^{-2}\cdot$ sec^{-1}, is $\alpha=(5.3\pm2.6)\times10^3$ at. units.

b. Change in the ionization probability at resonance. Experimental data on the resonance ionization process shown in Fig. 7 indicate that the ion-signal amplitude is proportional to the radiation intensity (Fig. 9).

c. Width of resonances. The results of the present experiment (Fig. 7) do not enable us to elucidate the dependence of the width of resonance states on the field strength. This is so because the difference between the resonance widths is of the same order as the measurement error in the width of the resonance maximum and the error in the measured instrumental factors. Estimates have shown that, at most, the change in the width is of the order of 0.1 cm^{-1}. Apart from the above width of the 2^1S–6^1S transition, the width of the observed resonances is also determined by instrumental factors such as inhomogeneity and Doppler broadening and the reduced linewidth of the laser radiation. Let us estimate these factors. Doppler broadening at room temperature is ≈0.2 cm^{-1}. The reduced linewidth of the laser radiation for the two-photon resonance was determined from the formula $\overline{\Gamma}_l=\sqrt{k}\Gamma_l$ for multimode radiation with random phase distribution (k is the degree of nonlinearity of the resonance transition and Γ_l is the linewidth of the radiation). It was found to be approximately 0.3 cm^{-1}. The broadening due to the inhomogeneity in the spatial distribution of the laser radiation over the target was calculated from the experimental data on the spatial distribution of the radiation intensity; it was found to be $\Gamma_l\approx0.6$ cm^{-1}.

It is also important to take into account the fact that the laser used in these experiments was working in the multimode state. It is well known that, when the width of the exciting Gaussian spectrum is comparable with the width of the atomic line (this was the case in our experiments), the absorption linewidth differs from the shape of the atomic line, and the shift of the maximum and the broadening are comparable with the width of the atomic line, [12] i.e., ≈0.1 cm^{-1}. The resultant width due to these independent factors is $\Gamma_\Sigma\approx0.6$ cm^{-1}.

The experiment has thus yielded the following results: 1) the change in the energy of the 2^1S–6^1S transition is a quadratic function of the field strength; 2) the constant corresponding to this dependence is $\alpha=(5.3\pm2.6)\times10^3$ at. units; 3) the change in the ionization probability at resonance is a quadratic function of the field. The experimental results taken altogether enable us to elucidate the nature of the resonance ionization process. As noted above, three-photon ionization in the presence of a two-photon intermediate resonance has particular features which distinguish it from other possible cases of resonance ionization. [11] Thus, when the degree of nonlinearity of the 2^1S–6^1S transition is $k(2^1S-6^1S)=2$ and the degree of nonlinearity of the transition from the 6^1S state to the continuum is $k(6^1S-E)=1$, the ionization and field widths are proportional to the square of the field and, therefore, the competition between the resonance and nonresonance perturbations is determined by the numerical parameters. When two-photon mixing of resonance states predominates, the ionization process should be described by the formula (24) given in [13]; when ionization from the 6^1S state predominates, the resonance formula given by Keldysh [14] is valid. The general solution for the resonance ionization of atoms, obtained in [15], has provided a qualitative confirmation of the validity of the solutions for the limiting cases discussed in [13] and [14]. However, it is not obvious that the results reported in [15] can be justifiably used for quantitative estimates in our intermediate case, since it was assumed in [15] that the field was turned on instantaneously whilst, in the experiment, this is an adiabatic process.

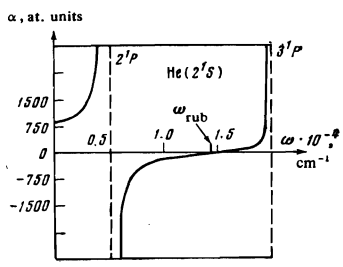

FIG. 10. Dynamic polarizability α of the helium atom in the $2\,^1S$ state as a function of frequency (theoretical calculation from [11,12]). ω_{rub} is the ruby laser frequency.

Let us estimate the numerical parameters that could be compared with the experimental results. Since the measured change in the energy of the 2^1S-6^1S transition (≈ 10 cm^{-1}) is much less than the distance to the nearest dipole-bound levels (≈ 90 cm^{-1}), we can use perturbation theory for our estimates. The change in the energy of the 2^1S-6^1S transition is determined by the difference between the dynamic polarizabilities of the resonance 2^1S and 6^1S states:

$$\Delta E(\mathscr{E}) = -\tfrac{1}{4}\{\mathrm{Re}\,\alpha(6^1S) - \mathrm{Re}\,\alpha(2^1S)\}\mathscr{E}^2.$$

Calculations of the dynamic polarizabilities of the helium atom in these states by the model-potential method[16,17] yield:

$$\alpha(2^1S) = -43.7 \text{ at. units}; \quad \alpha(6^1S) = (-214+7i) \text{ at. units}.$$

The use of wave functions obtained by other methods for the 2^1S state has practically no effect on $\alpha(2^1S)$. This quantity is small because the frequency $\omega \approx 14\,400$ cm^{-1} is close to the center of the interresonance $2^1P_0-3^1P_0$ interval in which the polarizability changes sign (Fig. 10). The single-photon ionization channel is open for the 6^1S state, and $\mathrm{Re}\,\alpha(6^1S)$ is not very different from the asymptotic value corresponding to the change in the energy by an amount equal to the mean kinetic energy of a free electron in the wave. Calculations of the polarizabilities show that the separation between the 2^1S and 6^1S levels is increased in the field by an amount proportional to $\mathscr{E}^2$, which is in agreement with experiment. However, the calculated value of α is lower by one and a half orders of magnitude as compared with the experimental value. The reason for this discrepancy is not as yet clear.

Ionization broadening of the resonance state can be determined from the imaginary part of the dynamic polarizability of the 6^1S state [$\mathrm{Im}\,\alpha(6^1S) = 7$ at. units] and from the photoionization cross section σ for the state ($\sigma = 4.9\times10^{-19}$ cm^2) obtained by another method.[18] In both cases, when the field is $\mathscr{E} \approx 5\times10^5$ V·cm^{-1}, we have $\Gamma_i(6^1S) \approx 0.1$ cm^{-1}, which is not inconsistent with experimental data and indicates that the widths of the observed resonances are determined by the instrumental factors (Γ_Σ), and are practically independent of the radiation intensity. The magnitude of Γ_i also shows that, when the laser pulse length is $\tau_p \approx 3\times10^{-8}$ sec, the transition from the resonance state to the continuum is saturated, i.e.,

$$\int_0^p W(6^1S-E)\,dt \approx 1$$

[$W(6^1S-E)$ is the probability of transition from the 6^1S state to the continuum]. When the 6^1S-E transition is saturated, the probability $W(6^1S-E)$ is independent of the field. The dependence of the resonance amplitude (probability of resonance ionization) on the field is then determined by the probability of populating the 6^1S state. If we estimate the probability of excitation at the exact resonance in the usual way, we find that

$$W(2^1S-6^1S) = |K(2^1S-6^1S)|^2/\Gamma,$$

where $K(2^1S-6^1S)$ is the composite transition matrix element and Γ is the natural width of the 6^1S level, which is determined from the imaginary part of the dynamic polarizability of the 6^1S state, and is given by $\Gamma = \frac{1}{2}\,\mathrm{Im}\,\alpha(6^1S)\mathscr{E}^2$.[16] Calculations of the matrix element $K(2^1S-6^1S)$[17] have shown that it is equal to $3.5\mathscr{E}^2$, where $\mathscr{E}$ is measured in atomic units. Thus, substituting for Γ and $K(2^1S-6^1S)$ into the expression for the ionization probability at resonance, we find that the resonance amplitude is proportional to the radiation intensity, which is in agreement with experiment (Fig. 9). Using the above value for the matrix element $K(2^1S-6^1S)$, we find that when $\mathscr{E} \approx 5\times10^5$ V·cm^{-1}, the excitation probability is far from saturation, i.e.,

$$\int_0^p W(2^1S-6^1S)\,dt \ll 1,$$

so that the mixing of the 2^1S and 6^1S states plays no appreciable role. The assumption that the resonance transition is saturated ensures that the ion-yield amplitude at resonance is independent of the intensity F, but this is not in agreement with experiment. Our analysis thus shows that, in this case, we have nonresonance perturbation of states by the field.

The authors are grateful to Prof. M. S. Rabinovich and Prof. L. P. Rapoport for valuable discussions, and to V. A. Grinchuk, G. A. Delone, V. G. Ovchinnikov, D. Rubin, and M. V. Fedorov for their help in this research.

[1)]Central Institute for Physical Studies, Hungarian Academy of Sciences.
[2)]Voronezh State University.

[1]N. B. Delone, Usp. Fiz. Nauk 115, 361 (1975) [Sov. Phys. Usp. 18, 169 (1975)].
[2]S. É. Frish, Sb. Spektroskopiya gazorazryadnoĭ plazmy (in: Spectroscopy of Gas Discharge Plasma), Nauka, 1970, p. 7.
[3]A. V. Phelps, Phys. Rev. 99, 1307 (1955).
[4]Yu. G. Kozlov, Opt. Spektrosk. 34, 34 (1968).
[5]B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. 61, 968 (1971) [Sov. Phys. JETP 34, 515 (1972)].
[6]G. A. Delone, Kratk. Soobshch. Fiz. No. 8 (1975); G. A. Delone, N. L. Manakov, M. A. Preobrazhenskiĭ, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. 70, 1234 (1976) [Sov. Phys. JETP 43, 642 (1976)].
[7]N. Isenor and M. Gervenan, Opt. Commun. 10, 280 (1974).
[8]G. A. Delone, N. B. Delone, V. K. Zolotarev, N. L. Manakov, G. K. Piskova, and M. A. Tursunov, Zh. Eksp. Teor. Fiz. 65, 481 (1973) [Sov. Phys. JETP 38, 236 (1974)].
[9]V. A. Grinchuk, G. A. Delone, and K. B. Petrosyan, Fiz. Plazmy 1, 321 (1975); V. A. Grinchuk and K. B. Petrosyan, Kratk. Soobshch. Fiz. No. 1 (1975); V. A. Grinchuk, G. A. Delone, and K. B. Petrosyan, Kratk. Soobshch. Fiz. No. 3 (1975); G. A. Delone, B. A. Zon, and K. B. Petrosyan, Pis'ma Zh. Eksp. Teor. Fiz. 22, 519 (1975) [JETP Lett. 22, 253 (1975)].
[10]J. Bakos, A. Kiss, L. Szabo, and M. Tendler, Phys. Lett. A 39, 283 (1972); 39, 317 (1972); 41, 163 (1972); J. Bakos, A. Kiss, L. Sabo, and M. Tendler, Pis'ma Zh. Eksp. Teor. Fiz. 18, 403 (1973) [JETP Lett. 18, 237 (1973)].
[11]N. B. Delone, Sb. Avtoionizatsionnye yavleniya v atomakh

(in: Autoionization Phenomena in Atoms), Moscow State University, 1976.
[12]A. M. Bonch-Bruevich, S. G. Przhibel'skiĭ, V. A. Khodovoĭ, and N. A. Chigir', Zh. Eksp. Teor. Fiz. 70, 445 (1976) [Sov. Phys. JETP 43, 230 (1976)].
[13]L. P. Kotova and M. V. Terent'ev, Zh. Eksp. Teor. Fiz. 52, 732 (1967) [Sov. Phys. JETP 25, 481 (1967)].
[14]L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov. Phys. JETP 20, 1307 (1965)].
[15]L. Armstrong and B. L. Beers, Phys. Rev. A 12, 1903 (1975); A. E. Kazakov, V. P. Makarov, and M. V. Fedorov, Zh. Eksp. Teor. Fiz. 70, 38 (1976) [Sov. Phys. JETP 43, 20 (1976)].
[16]N. L. Malakov, V. D. Ovsyannikov, and L. P. Rapoport, Opt. Spektrosk. 38, 206 (1975).
[17]N. L. Malakov, V. D. Ovsyannikov, and L. P. Rapoport, Zh. Ekxp. Teor. Fiz. 70, 1697 (1976) [Sov. Phys. JETP 43, (1976)].
[18]L. A. Vaĭnshteĭn and V. P. Shevel'ko, Preprint No. 26, Lebedev Physics Institute, Moscow, 1974.

Translated by S. Chomet

Hydrogenlike system in crossed electric and magnetic fields

L. A. Burkova, I. E. Dzyaloshinskiĭ, G. F. Drukarev, and B. S. Monozon

Leningrad State University
(Submitted March 4, 1976)
Zh. Eksp. Teor. Fiz. **71**, 526–530 (August 1976)

The problem of a hydrogenlike system in crossed electric and magnetic fields is investigated. A quasiclassical calculation is made of the energy of a state in which the wave function is concentrated at a certain distance from the center of the Coulomb well. The nature of the spectrum is ascertained and a criterion for the validity of the obtained results is established. A discussion is given of the conditions under which observation of the investigated states becomes feasible.

PACS numbers: 03.65.Sq, 03.65.Ge

1. INTRODUCTION

A hydrogenlike system (hydrogen atom, exciton) on which crossed electric and magnetic fields act is investigated in this article. The center of inertia is fixed. We shall assume that $\mathscr{E} \ll \mathscr{H}$. This assumption allows us to confine our attention to a nonrelativistic treatment. The posed problem is physically equivalent to the problem of a particle moving with velocity $\mathbf{V}$ in a homogeneous magnetic field $\mathscr{H}$, since an electric field

$$\mathscr{E} = \frac{1}{c}[\mathbf{V} \times \mathscr{H}] \tag{1}$$

appears in the coordinate system attached to the particle.

Choosing the symmetric gauge of the vector potential

$$\mathbf{A} = {}^1/_2[\mathscr{H} \times \mathbf{r}] \tag{2}$$

($\mathbf{r}$ denotes the distance between the positively charged and negatively charged particles), we obtain the Schrödinger equation for the problem under consideration in the form[1]

$$\left\{-\frac{\hbar^2}{2\mu}\Delta + \frac{ie\hbar}{2\mu c}\gamma\mathscr{H}[\mathbf{r}\times\nabla] + \frac{e^2}{8\mu c^2}[\mathscr{H}\times\mathbf{r}]^2 + e\mathscr{E}\mathbf{r} - \frac{e^2}{r}\right\}\Psi = E\Psi, \tag{3}$$

where ε is the dielectric constant ($\varepsilon = 1$ for the hydrogen atom),

$$\mu = \frac{m_+m_-}{m_+ + m_-}, \quad \gamma = \frac{m_+ - m_-}{m_+ + m_-}, \tag{4}$$

m_+ and m_- denote the masses of the positively and negatively charged particles.

Following Gor'kov and one of the authors,[1] let us set

$$\Psi = \Phi \exp\left\{i\gamma M \frac{c}{2\hbar\mathscr{H}^2}[\mathscr{H}\times\mathscr{E}]\mathbf{r}\right\}, \tag{5}$$

where $M = m_+ + m_-$. Having directed the z axis along the magnetic field, the y axis along the electric field, denoting the radius vector in the (x, y) plane by ρ, and displacing the origin of coordinates along the y axis by the amount

$$y_0 = -Mc^2\mathscr{E}/e\mathscr{H}^2, \tag{6}$$

we obtain the following equation for Φ[1]:

$$\left\{-\frac{\hbar^2}{2\mu}\Delta_\rho - \frac{\hbar^2}{2\mu}\frac{\partial^2}{\partial z^2} + \frac{ie\hbar\gamma}{2\mu c}[\rho\times\nabla_\rho]\mathscr{H} + \frac{e^2}{8\mu c^2}\mathscr{H}^2\rho^2 - \frac{e^2}{\varepsilon[x^2+(y+y_0)^2+z^2]^{1/2}} - \frac{Mc^2\mathscr{E}^2}{2\mathscr{H}^2}\right\}\Phi = E\Phi. \tag{7}$$

If we divide both sides of Eq. (7) by the quantity e^2, then three parameters having the dimension of a length are formed in the equation: the Bohr radius $a = \hbar^2/\mu e^2$, the magnetic radius $a_H = (\hbar c/e\mathscr{H})^{1/2}$, and the characteristic length y_0. Two, independent, dimensionless parameters can be constructed from these quantities

$$\lambda = y_0/a, \quad \nu = a_H/a. \tag{8}$$

In order to express λ and ν in terms of the field strength in the most convenient way, let us introduce the charac-