Three-photon ionization of metastable helium atoms

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Direct and resonance three-photon ionization processes are investigated for helium atoms in 2S and 2S 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 2S and 2S 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 radiation intensity, has been found for the ionization process in the presence of the two-photon intermediate 2S-6S resonance. The origin of perturbations of resonance states is discussed.

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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. 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. The field strength must be high and the adiabatic parameter low. 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. 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 $\gamma$ 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 ionization of helium atoms.
1. Target

Target is that it should contain the maximum density of atoms in metastable states. We have used the optimum discharge, all the highly excited states in the plasma passed through the tube and could be cut off in a time of about 1 Torr and discharge-tube diameter 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.

2. FORMULATION OF EXPERIMENT

The ionization energies of helium atoms in metastable states are \( E(2S) = 3.98 \text{ eV} \) and \( E(2S) = 4.77 \text{ eV} \). Using a comparison between the spectra of bound electronic states of the helium atom and energy of the radiation emitted by the ruby laser \( h\nu = 1.78 \text{ eV} \), and the selection rules for multiphoton transitions, one can show that ionization from the \( 2S \) state by plane and circularly-polarized radiation, and from the \( 2S \) 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 \( 6S \) state by varying the ruby-laser frequency during ionization from the \( 2S \) 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. 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 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 $10^5$. 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 $\xi$ 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 $\xi < 10^5$ 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 $\xi$ was varied between $2 \times 10^2$ and $5 \times 10^6$ V cm$^{-1}$, we determined the probe signal amplitude $A_i$ as a function of the energy $Q$ per pulse of the laser radiation, which was $10^3$ ions formed in the plasma. The results are shown in Fig. 1. The experiments were performed at frequencies in the range $\omega = 14407 - 14416$ 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 $k = 8 \log A/8 \log Q$, where $A_i$ is the probe signal amplitude per pulse of the laser radiation, which was $10^3$ ions formed in the plasma. The experiments were performed at frequencies in the range $\omega = 14407 - 14420$ cm$^{-1}$, where, if the above assumption is correct, the ionization of atoms should be a direct process. Figure 3 shows a typical result of an experiment of this kind. The results of such measurements enable 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 $\omega = 14415 - 14420$ cm$^{-1}$ whereas ionization from the triplet state predominates at frequencies in the range $14407 - 14410$ cm$^{-1}$.

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 $\tau_d$ of the laser pulse relative to the discharge-quenching pulse for constant energy per laser pulse at different frequencies $\omega$ in the range $\omega = 14407 - 14420$ cm$^{-1}$. Figure 4 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 $\omega = 14415 - 14420$ cm$^{-1}$ whereas ionization from the triplet state predominates at frequencies in the range $14407 - 14410$ 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:

\begin{align*}
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\end{align*}
\( | E_i - E_f| > \hbar \omega \), \( \gamma_i(\omega) \), \( \gamma_f(\omega) \),

where \( E_{0,1} \) is the energy of the bound electronic states in the atomic spectrum, \( \gamma_i \) is the reduced width for the 0, i transition, and \( \delta E_{0,1}(\omega) \) and \( \delta E_{0,i}(\omega) \) 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 \( \omega \) for fixed light intensity \( F \). Figure 4 shows the probe-signal amplitude \( A \) as a function of frequency \( \omega \) 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 \( \omega = 14420 \) cm\(^{-1} \), at which the ionization probability rapidly increases. Similar measurements performed for the ionization from the 2\( S \) 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\( S \) and 2\( S \) states by linearly and circularly polarized light is, in fact, a direct process of frequencies in the range \( \omega = 14402 - 14416 \) 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

\[
W(\omega) = 2\pi \left( \frac{F}{F_0} \right) \left\{ \frac{1}{27} \left[ T_{S0} + \frac{4}{5} T_{S1} \right] + \frac{4}{175} T_{S2} \right\},
\]

whereas, in the case of circularly-polarized radiation,

\[
W_c(\omega) = 2\pi \left( \frac{F}{F_0} \right) \left\{ \frac{2}{3} T_{S0} \right\}.
\]

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, \( T_{S0} \), is the radial composite matrix element expressed in terms of the radial Green function \( g_i(E; r, r') \) of the optical electron in the atom

\[
T_{S0} = \langle R | r g_i(E_{i}; r_0, r) r g_i(E_{f}; r_0, r) | 0 \rangle,
\]

and \( | i \rangle \) and \( | R_f \rangle \) are the wave functions of the initial and final states of the optical electron.

Numerical calculations of the function \( W_{123}(\omega) \) are shown in Fig. 5 for one interval between resonances (3\( P_{2} - 4\)S\(_2\) and 5\( D_{2} - 6\)S\(_2\)), which contains the ruby-laser frequency. The mean difference between \( W(2\)S\(_2\)) and \( W(2\)S\(_2\)) amounts to one or two orders [including the regions near the resonance and frequencies for which \( W(2;\omega) = 0 \)], which is to be expected in view of the substantial difference between the structures of the singlet and triplet terms. The substantial increase in \( W(2\)S\(_2\)) and \( W(2\)S\(_2\)), in comparison with \( W(2\)S\(_2\)) near \( \omega = 14400 \) cm\(^{-1} \), is connected with the proximity of the two-photon intermediate resonances 2\( S_{2} - 6\)S\(_2\) and 2\( S_{2} - 6\)D\(_2\) for which the detuning is about 30 and about 180 cm\(^{-1} \), respectively.

To calculate \( T_{S0} \), we used the wave functions for the initial 2\( S \) and 2\( S \) states, constructed in the quantum-mechanical defect approximation (QMD) and the Hartree-Fock method, whereas the Green function \( g_i \) was obtained in the QMD approximation.(6) 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\( S \) state, but plays a minor role in the case of ionization from the 2\( S \) 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-
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 $\tau_2 \geq 100$ ms, the dominant process is ionization from the $2^3S$ state whereas, for $50 \leq \tau_2 \leq 100$ ms, the contributions of ionization from $2^3S$ and $2^1S$ 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 $A(2^3S)/A(2^1S)$ and $n_0(2^3S)/n_0(2^1S)$ enable us to conclude that, at $\omega = 14.412$ cm$^{-1}$ and for plane-polarized light, the cross-section ratio is \( \alpha(2^3S)/\alpha(2^1S) = 10 \pm 5.3 \). 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 $^{27,28}$. 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 $A_1/A_2$ of probe signals at each fixed radiation frequency is equal to the cross section ratio $\alpha_1/\alpha_2$ for linear (l) and circular (c) polarizations. We have obtained the following results: $\alpha_2/\alpha_1 = 0.5 \pm 0.15 (2^3S)$ for frequencies $\omega$ between 14.412 and 14.415 cm$^{-1}$ and $\alpha_2/\alpha_1 = 0.35 \pm 0.15 (2^3S)$ at $\omega = 14.407$ 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 $2^3S$ 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 $2^3S$ state are determined by the “resonance” matrix element $T_{10}$, and the use of the Hartree–Fock wave function for the $2^3S$ 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 “dephasing” 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 $\langle n^1S | r | n^1S \rangle$ with approximate functions $| n^1S \rangle$ and $| n^1S \rangle$ 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 $2^3S$ state by plane-polarized radiation of frequency $\omega = 14.415$ cm$^{-1}$. It is known$^{17}$ that the cross section $\sigma_3$ for three-photon ionization is related to the measured parameters by the formula

\[
\sigma_3 = \frac{N_I}{n_0} \left( \frac{\omega}{Q} \right) \frac{S^4}{V_\tau} \tag{4}
\]

where $N_I$ is the number of ions produced by the laser radiation, $n_0$ is the neutral atom density, $Q$ and $\omega$ are, respectively, the energy per pulse and radiation frequency,

\[
V_\tau = \int f(x, y, z) dx, \quad \tau_1 - \int \psi'(t) dt
\]

are the volume of the target and the time of interaction.

\[
\text{FIG. 6. Probe-signal amplitude } A_1 \text{ as a function of the delay of the laser pulse relative to the quenched pulse at the frequency } \omega = 14.412 \text{ cm}^{-1}.
\]
with the field involving the absorption of three photons in a single event, \( f(x, y, z) \) and \( \phi(t) \) are the spatial and temporal distributions of the radiation intensity, and \( S \) and \( T \) 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 \( \alpha \) can be indicated. For plane-polarized radiation, \( \alpha = 10^{-14} \text{ cm}^3 \cdot \text{sec}^2 \). 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 \( 2S \) 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. 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 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 \( k_{s0} > 2k_{sE} \), where \( v \) 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. The resonance that can be produced during the ionization of the helium atom from the \( 2S \) state has a different character, since \( k(2S-6S) = 2k(6S-E) = 2 \). 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 \( 2S \) and \( 6S \) states predominates in this case.

Measurements of the probability \( W(2S) \) 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 \( 2S-6S \) 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 \( 2S-6S \) transition undergoes a substantial change in the light field. It is clear from Fig. 4 that the resonance ion yield corresponds to \( \omega = 14420.7 \text{ cm}^{-1} \) which differs by about \( 2 \text{ cm}^{-1} \) from the resonance frequency \( \omega_0 = [E(6S) - E(2S)]/2\epsilon = 14418.7 \text{ cm}^{-1} \) 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 (= 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 \( \pm 0.1 \text{ cm}^{-1} \).

a. Change in the energy of the \( 2S-6S \) transition.

The dependence of the \( 2S-6S \) 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
dence of the change in the transition energy on the field strength: \( \Delta E(6^1S-2^1S) = \alpha^2 \). To determine the absolute magnitude of the coefficient \( \alpha \), 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 \( F_{\text{eff}} = 0.55 F_{\text{max}} \). The value of \( \alpha \) for the \( 2^1S-6^1S \) transition, determined from experimental data on \( \Delta E \) for the corresponding \( F_{\text{eff}} = 6.6 \times 10^8 \text{ cm}^{-2} \cdot \text{sec}^{-1} \), is \( \alpha = (5.3 \pm 2.6) \times 10^3 \) 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 \( 0.2 \text{ cm}^{-1} \). The reduced linewidth of the laser radiation for the two-photon resonance was determined from the formula \( \Gamma_0 = \sqrt{k} \Gamma_1 \) for multimode radiation with random phase distribution \( k \) is the degree of nonlinearity of the resonance transition and \( \Gamma_1 \) is the linewidth of the radiation). It was found to be approximately \( 0.3 \text{ cm}^{-1} \). 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 \( \Gamma_1 = 0.6 \text{ cm}^{-1} \).

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, \( \Gamma_0 = 0.1 \text{ cm}^{-1} \). The resultant width due to these independent factors is \( \Gamma_0 = 0.6 \text{ cm}^{-1} \).

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 \( \alpha = (5.3 \pm 2.6) \times 10^3 \) 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. 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 \( ^{11} \); 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 \( ^{11} \), 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 \( ^{11} \) can be justifiably used for quantitative estimates in our intermediate case, since it was assumed in \( ^{11} \) that the field was turned on instantaneously whilst, in the experiment, this is an adiabatic process.

![Fig. 8](image8.png)

FIG. 8. Change \( \Delta E \) of the \( 2^1S-6^1S \) transition energy as a function of the radiation intensity \( F \).

![Fig. 9](image9.png)

FIG. 9. Probe-signal amplitude at resonance \( A_{\text{max}} \) as a function of the radiation intensity \( F \).

![Fig. 10](image10.png)

FIG. 10. Dynamic polarizability \( \alpha \) of the helium atom in the \( 2^1S \) state as a function of frequency (theoretical calculation from \( ^{11,15} \)). \( \omega_{\text{res}} \) 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 $2^3S - 6^3S$ transition ($= 10 \text{ cm}^{-1}$) is much less than the distance to the nearest dipole-bound levels ($= 90 \text{ cm}^{-1}$), we can use perturbation theory for our estimates. The change in the energy of the $2^3S - 6^3S$ transition is determined by the difference between the dynamic polarizabilities of the resonance $2^3S$ and $6^3S$ states:

$$\Delta E(\sigma) = -V, (\text{Re} \alpha(2^3S) - \text{Re} \alpha(6^3S)) \sigma^2.$$  

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

$$\alpha(2^3S) = -43.7 \text{ at. units}; \alpha(6^3S) = (-214 + 7i) \text{ at. units}.$$  

The use of wave functions obtained by other methods for the $2^3S$ state has practically no effect on $\alpha(2^3S)$. This quantity is small because the frequency $\omega = 14000 \text{ cm}^{-1}$ is close to the interresonance $2^3P_2 - 3^3P_2$ interval in which the polarizability changes sign (Fig. 10). The single-photon ionization channel is open for the $6^3S$ state, and $\text{Re} \alpha(6^3S)$ 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 $2^3S$ and $6^3S$ levels is increased in the field by an amount proportional to $\sigma^2$, which is in agreement with experiment. However, the calculated value of $\alpha$ 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 $6^3S$ state [$\text{Im} \alpha(6^3S) = 7 \text{ at. units}$] and from the photoionization cross section $\sigma$ for the state ($\sigma = 4.9 \times 10^{-14} \text{ cm}^2$) obtained by another method.\textsuperscript{[18]} In both cases, when the field is $\sigma = 5 \times 10^{13} \text{ V cm}^{-1}$, we have $\Gamma(6^3S) = 0.1 \text{ cm}^{-1}$, which is not inconsistent with experimental data and indicates that the widths of the observed resonances are determined by the instrumental factors ($\Gamma_G$), and are practically independent of the radiation intensity. The magnitude of $\Gamma_G$ also shows that, when the laser pulse length is $\tau_L = 3 \times 10^{-8} \text{ sec}$, the transition from the resonance state to the continuum is saturated, i.e.,

$$\int_0^\infty W(6^3S - E) \text{d}E = 1,$$

$W(6^3S - E)$ is the probability of transition from the $6^3S$ state to the continuum. When the $6^3S - E$ transition is saturated, the probability $W(6^3S - E)$ 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 $6^3S$ state. If we estimate the probability of excitation at the exact resonance in the usual way, we find that

$$W(2^3S - 6^3S) = |K(2^3S - 6^3S)| \Gamma/\tau,$$

where $K(2^3S - 6^3S)$ is the composite transition matrix element and $\Gamma$ is the natural width of the $6^3S$ level, which is determined from the imaginary part of the dynamic polarizability of the $6^3S$ state, and is given by $\Gamma = \frac{1}{2} \text{Im} \alpha(6^3S) \sigma^2.$ Calculations of the matrix element $K(2^3S - 6^3S)$ have shown that it is equal to $3.5 \times 10^{-6}$, where $\sigma$ is measured in atomic units. Thus, substituting for $\Gamma$ and $K(2^3S - 6^3S)$ 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 $K(2^3S - 6^3S)$, we find that when $\sigma = 5 \times 10^{13} \text{ V cm}^{-1}$, the excitation probability is far from saturation, i.e.,

$$\int_0^\infty W(2^3S - 6^3S) \text{d}E \approx 1.$$  

so that the mixing of the $2^3S$ and $6^3S$ 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.

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\textsuperscript{14}N. B. Delone, Sb. Avtoionizatsionnye yavleniya v atomakh
Hydrogenlike system in crossed electric and magnetic fields

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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.

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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 \( \mathcal{E} \ll \mathcal{H} \). 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 \( \mathcal{H} \), since an electric field \( \mathbf{E} = -\frac{\mathbf{V} \times \mathcal{H}}{c} \) appears in the coordinate system attached to the particle.

Choosing the symmetric gauge of the vector potential

\[
\mathbf{A} = \frac{1}{2} \mathbf{[\mathcal{H} \times r]} \tag{2}
\]

(\( r \) denotes the distance between the positively charged and negatively charged particles), we obtain the Schrödinger equation for the problem under consideration in the form \( \psi = E \psi \).

\[
\begin{align*}
\left\{ \frac{\hbar^2}{2\mu} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} \right) + \frac{e^2}{8\mu \epsilon} \left[ \mathcal{H} \times \mathbf{r} \right]^2 + \frac{e^2}{\epsilon} \right\} \psi = E \psi, \tag{3}
\end{align*}
\]

where \( \epsilon \) is the dielectric constant (\( \epsilon = 1 \) for the hydrogen atom),

\[
\begin{align*}
\mu &= \frac{m_+ m_-}{m_+ + m_-}, \\
\gamma &= \frac{m_- - m_+}{m_+ + m_-}.
\end{align*} \tag{4}
\]

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