At $I>I_0$ bound states appear in the standing-wave field and contribute to the fine structure of the absorption coefficient. At $I>I_1$ the general form of the absorption coefficient changes: the Doppler contour gives rise to an absorption band with a width proportional to the field amplitude.

The structure of the absorption peaks due to the bound states depends strongly on the standing-wave field parameters and on the width of the resonance. In the case of atomic resonances the peaks have a relatively simple form and their width is either equal to the resonance width or is of the order of the frequency detuning of the strong field (at small detunings). The narrow molecular resonances can form peaks of more complicated form. In particular, "quantization" of the peaks sets in: when the strong-field frequency is changed by an amount $\varepsilon(\hbar k)/\hbar$, a narrow band is detached from the peak.

When the condition (1) is satisfied, discontinuities can appear in the spectrum of the atoms and can lead to formation of dips in the absorption coefficient.

The appearance of narrow peaks or dips in the absorption coefficient is of interest for frequency standards. We note that although the position of the peak depends on the field intensity, the effect of splitting of one peak into two is determined only by the field detuning. The position of the dip does not depend on the field.

The authors thank S. G. Rautian and A. M. Shalagin for a useful discussion.

1)Hall[13] has reported observation (by an indirect method) of the recoil effect, as deduced from the shape of the Lamb dip in a laser with methane call. Direct observation is possible under the condition (1), which can be satisfied at low pressure and at large transit times (the diameter of the light beam is $\gtrsim 10$ cm). The critical intensity corresponding to this condition is $I_0 \lesssim 10^{-5}$ W/cm^2. The criterion (1) can be satisfied also for certain weakly-resolved atomic transitions, for example the intercombination transition $4^1S_0-4^3P_1$, $\lambda=6572$ Å, $I_0\sim 10^{-4}$ W/cm^2 in Ca.

2)If the criterion (1) is satisfied only for slow atoms, then the depth of the dip decreases.

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Nonlinear Doppler-free narrow resonances in optical transitions and annihilation radiation of positronium

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The possibility of Doppler-free narrow resonances is considered for transitions between fine-structure levels of the ground and first excited states of the positronium atom. An analysis is given of the conditions necessary for the observation of narrow saturation resonances in the case of single-quantum absorption in 1S-2P transitions, and narrow two-photon absorption resonances in 1S-2S transitions. It is shown that it is possible to obtain 2γ-annihilation lines from the positronium atom with widths much smaller than the Doppler width.

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1. INTRODUCTION. FORMULATION OF THE PROBLEM

Studies of the structure of the energy levels of the bound system consisting of an electron and a positron, i.e., the positronium (Ps) atom, and precise measurements of the transition frequencies of this atom, provide a unique possibility for a test of relativistic quantum theory. It is well known that there are two experimental methods at present for investigating the positronium energy levels, namely, the direct microwave method in which the fine-structure intervals are determined for the first excited state,[1] which provides a direct comparison between experiment and relativistic calculations, and the indirect method in which measurements are made on the $1\,^1S_0$ and $1\,^3S_1$ fine-structure interval in the ground state, which is based on a comparison between the experimentally observed quenching of the 3γ annihilation radiation in a magnetic field with theoretical predictions.[2]

Much more extensive information about the structure of the energy levels, and much more precise comparison between experiment and the predictions of relativistic theory, can be achieved by investigating the positronium atom by Doppler-free ultrahigh-resolution laser spectroscopy which, at present, results in the highest precision of spectroscopic measurements of transition frequencies of atoms and molecules in the gaseous state.[3] It is readily seen that the densities of positronium atoms are exceedingly low, so that spectroscopic studies based on the usual methods of detection cannot be employed either for laser absorption or optical fluorescence of excited states. In the case of the positronium atom, measurements of optical excitation effects must be based, as in microwave experiments, either on the counting of annihilation gamma rays or, when the frequency of the laser radiation differs from the frequency of the spontaneously emitted optical photon, on the counting of coincidences (anti-coincidences) between annihilation gamma rays and optical photons.

In addition to measurements of the frequencies of transitions between fine-structure levels in the ground and excited states of positronium, there is considerable independent interest in the possibility noted in[4] of producing narrow and frequency-tunable resonances in 2γ-annihilation emission of the positronium atom when laser radiation acts on an inhomogeneously-broadened optical transition line.

In this paper, we examine the possibility of a precise determination of the frequencies of optical transitions in the positronium atom by the methods of Doppler-free laser spectroscopy, and the possible control of the 2γ-annihilation spectrum by coherent laser radiation.

2. NARROW SATURATION RESONANCES IN SINGLE-PHOTON ABSORPTION IN 1*S*-2*P* TRANSITIONS

The saturated absorption spectrum of the positronium atom in a standing laser-light wave ($\lambda = 2430$ Å) should contain four narrow L_α resonances corresponding to the $1\,^1S_0-2\,^1P_1$ transition in parapositronium (p-Ps) and the three $1\,^3S_1-2\,^3P_J$ transitions ($J = 0, 1, 2$) in orthopositronium (o-Ps).[5] The widths of all these resonances are mainly due to the contribution of spontaneous L_α radiation, and amount to $\Gamma_1 \approx (2\pi\tau_{L_\alpha})^{-1} \approx 50$ MHz for a Doppler width (full width at half height) $\Delta\omega_D = 2(\ln 2)^{1/2}u/\lambda \approx 700$ GHz, where, according to[6], the mean thermal velocity of the positronium atom is $u \approx 10^7$ cm/sec.

It is well known that the number of o-Ps atoms is greater by a factor of three than the number of p-Ps atoms and, moreover, the size of the o-Ps cloud produced by o-Ps atoms diffusing into vacuum when the target is exposed to the positron beam[5,6] is greater than the corresponding size of the p-Ps cloud by the factor $\tau_T^{(0)}/\tau_S^{(0)} \approx 1.1 \times 10^3$ ($\tau_S^{(0)}$ and $\tau_T^{(0)}$ are the lifetimes of the p-Ps and o-Ps atoms in the ground state). For example, for the above value of the mean thermal velocity, the size of the o-Ps cloud is $L_T^{(0)} = u\tau_T^{(0)} \approx 1.4$ cm, whereas that of the p-Ps cloud is only $L_S^{(0)} = u\tau_S^{(0)} \approx 12\ \mu$. The main interest, therefore, attaches to the Doppler-free laser spectroscopy of o-Ps atoms.

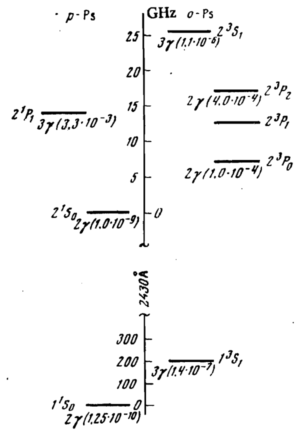

FIG. 1. Fine structure of the ground and first excited states, and the annihilation lifetime of the Ps atom.

Figure 1 shows the fine structure of the ground and the first excited states of the positronium atom, and indicates the lifetimes for the 2γ- and 3γ-annihilation decays.[7] The radiative-decay lifetimes of the fine-structure levels in the first excited state can readily be determined from the corresponding results for the hydrogen atom. The lifetime of the $2P$ levels is determined by L_α decay and is $\tau_{L_\alpha} \approx 3.2$ nsec, whereas, for the $2S$ levels, it is determined by the two-photon decay: $\tau_{2S} \approx 0.24$ sec. Since the annihilation lifetimes of the upper levels of all the $1S-2P$ transitions under consideration are different, it is possible to choose the most suitable method of detection of narrow saturation absorption resonances for each of them. Consider, for example, the detection of the narrow resonance corresponding to the $1\,^3S_1-2\,^3P_0$ transition. Since the upper level involved in this transition annihilates into two gamma rays (three-photon annihilation of o-Ps in the P state is strictly forbidden[7]), and the lower level annihilates into three gamma rays, the most suitable method of detection of the narrow optical resonance corresponding to this transition is that based on the reduction in the intensity of the 2γ-annihilation radiation from the $2\,^3P_0$ state as the laser frequency passes through the center of the transition line.

A possible experiment based on this method is illustrated schematically in Fig. 2. The o-Ps atoms, produced when the target 1 is exposed to a beam of slow positrons,[6] are excited by the standing laser wave (the target 1 is also used as a mirror), and the excited atoms are detected by recording the 2γ-annihilation signal with the gamma-ray counters 2. When the gamma-ray counters are used in coincidence, so that the only signal recorded is that corresponding to the coincidence of the two gamma rays, and the energy resolution of the system is such that the only pulses recorded

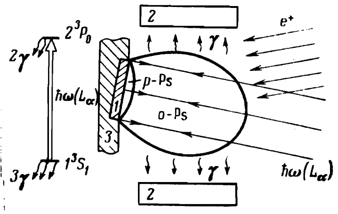

FIG. 2. Possible experiment involving the observation of the narrow resonance in the density of excited particles produced through the $1\,^3S_1-2\,^3P_0$ transition in the o-Ps atom; 1—target (mirror), 2—gamma-ray counters, 3—shield.

are those corresponding to gamma rays with energy $E_\gamma \approx 0.511$ MeV, the background signal will be determined by two factors, namely, the 2γ-annihilation radiation of o-Ps formed in the $2\,^3P_0$ and $2\,^3P_2$ states (it is shown in[8] that the fraction of these atoms is $\varepsilon \sim 10^{-4}$ of the total number of positronium atoms), and that part of the 3γ-annihilation radiation which contains two gamma rays with energies of about 0.511 MeV. The contributions of other sources of background gamma rays to the main 2γ-annihilation radiation of p-Ps can be made much smaller than the first two by a suitable choice of the geometry of the experiments and by the use of suitable upper and lower energy discriminators for the recorded gamma rays. In fact, if the geometry of the experiment is as shown in Fig. 2, only one gamma ray from the 2γ-annihilation radiation of p-Ps can directly enter the window of the gamma-ray counter and the second can only do so as a result of a Compton scatter through an angle $\sim 90°$. The energy of the second gamma ray is then ~ 0.255 MeV. The latter photon will not be recorded because of the energy discrimination in the system, and the coincidence signal due to the 2γ-annihilation radiation from p-Ps will not be recorded.

Let us now estimate the laser-flux density necessary to produce the saturated $1\,^3S_1-2\,^3P_0$ absorption. It is well known that the optimum value of the saturation parameter for the observation of the narrow density resonance corresponding to the excited particles is (we are assuming that the laser radiation is plane-polarized)[9]

$$G=\left(\frac{dE}{\hbar}\right)^2\frac{1}{\gamma_0(\gamma_1+\gamma)}\left(1+\frac{\gamma}{\gamma_0+\gamma_1}\right)^{-1}\approx 1, \tag{1}$$

where d is the dipole-moment matrix element for the transition under consideration, E is the laser field, $\gamma_0=1/\tau_T^{(0)}$, $\gamma_1=1/\tau_T^{(1)}$, $\gamma=1/\tau_{L_\alpha}$, $\tau_T^{(0)}\approx 139$ nsec, $\tau_T^{(1)}\approx 100$ μsec, and $\tau_{L_\alpha}\approx 3.2$ nsec (Fig. 1). The above expression for the saturation parameter was obtained as in[9], using approximate rate equations but, in contrast to[9], we took into account the spontaneous L_α emission between the upper and lower levels, which is important for the positronium atom.

If we use the general formulas for the matrix elements of the spherical components d_0, d_+, d_- of the vector **d** for atoms with LS coupling, [10] we can express the dipole-moment matrix element in terms of the reduced matrix element:

$$d=|\langle 2\,^3P_0|d_z|1\,^3S_1\rangle|=|\langle 2\,^3P_0|d_0|1\,^3S_1\rangle|={}^1/_3|\langle 2P\|d\|1S\rangle|. \tag{2}$$

Since, for the Ps atom, the radius of the first Bohr orbit is greater by a factor of two as compared with the case of the hydrogen atom, and if we use the value of the reduced matrix element for the hydrogen atom, [11] we can readily show that

$$d={}^2/_3 r_0 e R_{10}^{21}\approx 2.2D, \tag{3}$$

where r_0 is the Bohr radius and $R_{10}^{21}\approx 1.29$[10] is the overlap integral. Using (1) and (3) to eliminate E, we find that the laser-power density that is necessary to satisfy (1) is

$$I=\frac{cE^2}{8\pi}\approx\frac{c\hbar^2}{8\pi d^2}\gamma_0(\gamma_1+\gamma)\left(1+\frac{\gamma}{\gamma_0+\gamma_1}\right)\approx 2.7\ \text{W/cm}^2. \tag{4}$$

It is well known that when $G\approx 1$, i.e., for the above laser-power density, the depth of the valley in the density of the excited particles is less than the maximum by a factor of only two, and the broadening of the narrow resonance by the field is $\sqrt{2}$.[9]

From the experimental point of view, the main interest is in the time necessary for the detection of a narrow resonance for given signal-to-noise ratio (S/N):

$$\tau=(S/N)^2(i_b+i_u)/i_u^2, \tag{5}$$

where i_b and i_u are, respectively, the coincidence counting rates due to background gamma rays and the 2γ-annihilation photons from o-Ps atoms excited to the $2\,^3P_0$ state by the laser radiation. Suppose that N positronium atoms are produced in 1 sec when the target is illuminated, so that the stationary density of o-Ps atoms in the $1\,^3S_1$ state is $N_T^{(0)}=\frac{3}{4}N\tau_T^{(0)}$, and the numbers in the $2\,^3P_0$ and $2\,^3P_2$ states are $N_T^{(1)}=N_T^{(2)}=\frac{3}{4}\varepsilon N\tau_{L_\alpha}$. Correspondingly, the background counting rate due to the 2γ-annihilation radiation from o-Ps atoms formed in the $2\,^3P_0$ and $2\,^3P_2$ states, when the target is exposed to the positron beam, is

$$i_b'=\frac{3}{4}\varepsilon N\tau_{L_\alpha}\left(\frac{1}{\tau_T^{(1)}}+\frac{1}{\tau_T^{(2)}}\right)\frac{\Delta\Omega_\gamma}{4\pi}, \tag{6}$$

where $\tau_T^{(2)}\approx 400$ μsec is the lifetime of the $2\,^3P_2$ level for the 2γ-annihilation (we note that, for the $2\,^3P_1$ level, both the 2γ- and 3γ-annihilation radiation is strictly forbidden[7]) and $\Delta\Omega_\gamma/4\pi$ is the relative solid angle within which the gamma rays are recorded. Assuming that the discrimination system is such that only gammas with energies $mc^2(1\pm\delta)$ are recorded, the background counting rate due to the detection of two photons due to the 3γ-annihilation is

$$i_b''={}^3/_4 N\mu\Delta\Omega_\gamma/4\pi, \tag{7}$$

where the relative probability of emission of two gamma rays with energies $mc^2(1-\delta)\leq E_\gamma\leq mc^2$ in the case of 3γ-annihilation of o-Ps for $\delta\ll 1$ is[12]

$$\mu={}^5/_6\delta^2/(\pi^2-9). \tag{8}$$

It is clear from (6) and (7) that the main contribution to the background counting rate is provided by the second factor. For example, when $\delta=0.001$, [13] we have $i_b'/i_b''\approx 0.042$.

The laser radiation increases the density of o-Ps atoms in the $2\,^3P_0$ state by an amount[9]

$$\Delta N_T^{(1)}(\Omega)=N_T^{(0)}\frac{G}{1+\gamma_1/\gamma_0}f(G,\Omega), \tag{9}$$

where the function $f(G, \Omega)$ describes the narrow resonance valley in the density of atoms excited by the laser radiation at $\Omega=0$, where $\Omega=\omega-\omega_0$ is the detuning of the frequency ω of the laser radiation from the frequency ω_0 at the center of the line corresponding to the transition. When $G=1$, we have at the center of the line corresponding to the above transition

$$\Delta N_T^{(1)}(0)=\sqrt{\frac{\pi}{3}}N_T^{(0)}\frac{\Gamma_1}{ku}\frac{1}{1+\gamma_1/\gamma_0} \tag{10}$$

and the minimum useful signal is

$$i_u=\frac{\Delta N_T^{(1)}(0)}{\tau_T^{(1)}}\frac{\Delta\Omega_\gamma}{4\pi}=\frac{3}{4}\sqrt{\frac{\pi}{2}}N\frac{\Gamma_1}{ku}\frac{\tau_T^{(0)}}{\tau_T^{(0)}+\tau_T^{(1)}}\frac{\Delta\Omega_\gamma}{4\pi}, \tag{11}$$

where $ku\approx 420$ GHz. Substituting $S/N=3$, $\Delta\Omega_\gamma/4\pi=0.25$, $\varepsilon=10^{-4}$, $\delta=10^{-3}$, and the minimum time of detection $\tau=1$ h, we find from (5), (6), (7), and (11) that the necessary flux of positronium atoms is $N\approx 3\times10^5$ sec^{-1}. The total number of recorded background photons is then $i_b\tau\approx 190$, and the useful signal amounts to $i_u\tau\approx 40$ photons. We note that, in the experiments described in[1], which are concerned with measurements of the fine-structure intervals corresponding to the first excited state of the positronium atoms for a source activity of 500 μCi, the flux of the Ps atoms was 1.2×10^5 sec^{-1}. It is clear that, when the Ps flux and the signal-to-noise ratio S/N are fixed, the detection time will be much smaller for any other detuning of the laser frequency from the frequency at the center of the line corresponding to the transition.

We note that the recoil effect is important for the Ps atom.[14] Recoils ensure that each L_α resonance is split into two, and this splitting ($\Delta\approx 6.2$ GHz) is much greater than the homogeneous linewidth. The splitting is, of course, unimportant for the above estimate.

3. NARROW TWO-PHOTON 1S-2S RESONANCES

From the point of view of laser spectroscopy without the Doppler broadening of the Ps atom, the most interesting problem is the production of narrow nonlinear resonances in two-photon absorption[15] corresponding to the $1\,^1S_0$ and $2\,^1S_0$ transition in p-Ps, and the $1\,^3S_1-2\,^3S_1$ transition in o-Ps, which require laser radiation of wavelength $\lambda=4860$ Å. This problem is made much easier by the successful realization of narrow two-photon resonances, using the $1S-2S$ transition in hydrogen[17] proposed in[16].

Let us now consider the detection of the narrow two-photon absorption resonance in o-Ps. Since the $2\,^3S_1$ level is metastable against radiative decay, and the type of annihilation decay of the upper and lower levels is the same, the detection of the two-photon absorption resonance based on the change in the intensity of the 3γ-annihilation radiation is difficult. However, if the positronium atoms are located in a microwave field producing transitions from the $2\,^3S_1$ state to any one of the $2\,^3P_J$ states ($J=0, 1, 2$), or in a magnetic field that mixes $2\,^1S_0$ ($m=0$) and $2\,^3S_1$ ($m=0$) sublevels of para- and orthopositronium, the detection of the two-photon absorption resonance can be based either on coincidences between an L_α photon due to the $2\,^3P_J-1\,^3S_1$ transition and a gamma ray associated with the 3γ-annihilation radiation from the $1\,^3S_1$ state, or on the 2γ-annihilation of the mixed state $2\,^3S_1$ in a magnetic field.

Let us now determine the probability of two-photon excitation of o-Ps from the $1\,^3S_1$ to the $2\,^3S_1$ state in the standing light wave ($\lambda=4860$ Å). Taking into account only the simultaneous contributions of the two progessive waves, i.e., considering only the region near the center of the absorption line, it is found that the probability of two-photon excitation is[18]

$$W=\frac{2^4}{\hbar^2\Gamma}\left|\sum_r\frac{V_{2S,r}V_{r,1S}}{\hbar\omega-\hbar\omega_{r,1S}}\right|^2\frac{\Gamma_{2S}}{\Gamma_2}L(2\omega-\omega_{1S,2S}), \tag{12}$$

where

$$L(2\omega-\omega_{1S,2S})=\frac{\Gamma_2^2/4}{(2\omega-\omega_{1S,2S})^2+\Gamma_2^2/4}, \tag{13}$$

ω is the laser frequency, $\omega_{r,1S}$ is the frequency of the intermediate transition, $\omega_{1S,2S}$ and Γ_2 are the frequency and homogeneous width of the transition, respectively, and Γ_{2S} is the width of the upper $2\,^3S_1$ level. We shall suppose, and this is usually the case, that the laser radiation is plane-polarized, so that the composite matrix element is given by

$$M=\sum_r\frac{V_{2S,r}V_{r,1S}}{\hbar\omega-\hbar\omega_{r,1S}}=(eE)^2\sum_r\frac{z_{2S,r}z_{r,1S}}{\hbar\omega-\hbar\omega_{r,1S}}, \tag{14}$$

where z_{ba} are the matrix elements of the coordinate $z=z_--z_+$, and z_- and z_+ are the coordinates of the electron and positron in the positronium atom, respectively. Since only the space part of the wave function of the positronium atom is of interest for the evaluation of the matrix elements of the coordinate z, we shall use the corresponding results for the hydrogen atom[11] (fine structure of levels is neglected in the calculation):

$$z_{2S,r}=z_{2S,nP}=\frac{r_0}{\sqrt{3}}R_{20}^{n1},\qquad z_{r,1S}=z_{nP,1S}=\frac{r_0}{\sqrt{3}}R_{10}^{n1}, \tag{15}$$

$$\hbar\omega-\hbar\omega_{r,1S}=\hbar\omega-\hbar\omega_{nP,1S}=\mathrm{Ry}\cdot(5/8-1/n^2), \tag{16}$$

and the composite matrix element for hydrogen

$$M_H=\frac{(eEr_0)^2}{3\,\mathrm{Ry}}A, \tag{17}$$

where[1)]

$$A=\sum_{n=2}^{\infty}\frac{R_{10}^{n1}R_{20}^{n1}}{5/8-1/n^2}\approx 22, \tag{18}$$

and $R_{nl}^{n'l'}$ are the overlap integrals. For the positronium atom, we must substitute $r_0\to 2r_0$ and $Ry\to\frac{1}{2}Ry$ in (17), so that

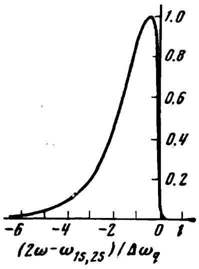

FIG. 3. Shape of resonance line corresponding to two-photon absorption. The line shape is normalized to unity at the maximum and is corrected for the quadratic Doppler effect.

$$M=\frac{2^3}{3}\frac{(eEr_0)^2}{\mathrm{Ry}}A \qquad (19)$$

and the probability of two-photon excitation of the $2\,^3S_1$ level is

$$W=\frac{2^{10}}{3^2}(eEr_0)^4\left(\frac{A}{\hbar\,\mathrm{Ry}\,\Gamma_2}\right)^2\Gamma_{2S}L(2\omega-\omega_{1S,2S}). \qquad (20)$$

Inclusion of the quadratic Doppler effect is important for the positronium atom. In fact, for example, for atoms with velocities of the order of the thermal velocity $u\approx 10^7$ cm/sec, the relative frequency shift due to the quadratic Doppler effect is $\Delta\omega/\omega=u^2/2c^2\approx 5.5\times 10^{-8}$, whereas the ratio of the homogeneous linewidth to the frequency of the $1\,^3S_1-2\,^3S_1$ transition is only $\Gamma_2/\omega\approx 5.2\times 10^{-10}$ ($\Gamma_2\approx 1.3$ MHz). If we now consider the probability of two-photon excitation due to only the simultaneous contribution of both progressive waves, we can take into account the quadratic Doppler effect by replacing 2ω by $2\omega+\omega v^2/c^2$ in (13), and then average the resulting expression over the Maxwellian velocity distribution $w(\mathbf{v})$. In that case,

$$\bar{L}(2\omega-\omega_{1S,2S})=\int L(2\omega-\omega_{1S,2S})\,w(\mathbf{v})\,d^3v=\frac{\Gamma_2}{2\Delta\omega_q}\mathrm{Re}\{iz^{1/2}\Psi(3/2,3/2,z)\}. \qquad (21)$$

where $\Delta\omega_q=\omega u^2/c^2\approx\omega_{1S,2S}u^2/2c^2$, $z=(2\omega-\omega_{1S,2S})/\Delta\omega_q+i\Gamma_2/2\Delta\omega_q$, and $\Psi(a,b,c)$ is a confluent hypergeometric function of the second kind. In the important case when $\Gamma_2/2\Delta\omega_q\ll 1$ (for the transition which we are considering $\Gamma_2/2\Delta\omega_q\approx 4.7\times 10^{-3}$), the expression for the line shape is substantially simplified and takes the form

$$\bar{L}(2\omega-\omega_{1S,2S})\approx\sqrt{2\pi}\frac{\Gamma_2}{2\Delta\omega_q}\exp\left\{\frac{2\omega-\omega_{1S,2S}}{\Delta\omega_q}\right\}$$
$$\times\left\{\left[\left(\frac{2\omega-\omega_{1S,2S}}{\Delta\omega_q}\right)^2+\left(\frac{\Gamma_2/2}{\Delta\omega_q}\right)^2\right]^{1/2}-\frac{2\omega-\omega_{1S,2S}}{\Delta\omega_q}\right\}^{1/2}, \qquad (22)$$

in which case, the maximum of the two-photon absorption resonance is red-shifted relative to the center of the atomic transition line by the amount

$$2\omega-\omega_{1S,2S}\approx-\tfrac{1}{2}\Delta\omega_q, \qquad (23)$$

and the maximum value is

$$\bar{L}\left(-\frac{\Delta\omega_q}{2}\right)=\sqrt{\frac{2\pi}{e}}\frac{\Gamma_2}{2\Delta\omega_q}. \qquad (24)$$

Accordingly, when $\Gamma_2/2\Delta\omega_q\ll 1$, the contrast of the resonance, i.e., the ratio of the maximum probability of two-photon excitation due to the simultaneous contribution of both progressive waves to the maximum probability of excitation due to the independent contributions of the two waves is $4\sqrt{2}ku/\sqrt{e}\Delta\omega_q$.

We note that, when $\Gamma_2/2\Delta\omega_q\ll 1$, the expression given by (22) is approximately valid for any detuning $|2\omega-\omega_{1S,2S}|\lesssim\Delta\omega_q$ and not only for $|2\omega-\omega_{1S,2S}|\ll\Delta\omega_q$.[19]

Figure 3 shows the line shape calculated from (22) for the two-photon absorption resonance corresponding to the $1\,^3S_1-2\,^3S_1$ transition in o-Ps. When $u\approx 10^7$ cm/sec, the resonance shift for this transition is $\Delta\omega_q/2\approx 68$ MHz, and the full width at half height is $\Delta\nu_q\approx 245$ MHz. The probability of two-photon excitation expressed in terms of the laser power density $I=cE^2/8\pi$ is, according to (20), then given by

$$\bar{W}=4.3\cdot 10^{-3}I^2\bar{L}(2\omega-\omega_{1S,2S}), \qquad (25)$$

where I is in W/cm^2.

Consider now the detection of the narrow two-photon absorption resonance based on the coincidences between L_α and γ photons associated with the $2\,^3S_1-2\,^3P_2$ transitions stimulated by the microwave field. We shall suppose that the microwave power is such that the probability that the positronium atom will undergo a transition under the action of this field is $W_C\gg W_T^{(3)}$, where $W_T^{(3)}\approx 0.91\times 10^6$ sec^{-1} is the probability of 3γ-annihilation of the excited state to $2\,^3S_1$. This condition ensures effective transfer of atoms from the $2\,^3S_1$ to the $2\,^3P_2$ state. Moreover, for simplicity, we shall suppose that $W_C\ll W_L\approx 3.1\times 10^8$ sec^{-1}. In the absence of the laser radiation, the populations of the $2\,^1P_1$, $2\,^3P_2$, $2\,^3P_1$, and $2\,^3P_0$ levels are then, respectively, equal to $\frac{3}{4}\varepsilon N\tau_{L\alpha}$, $\frac{3}{2}\varepsilon N\tau_{L\alpha}$, $\frac{3}{4}\varepsilon N\tau_{L\alpha}$, and $\frac{3}{4}\varepsilon N\tau_{L\alpha}$, and the background $L_\alpha-\gamma$ coincidence signal is

$$i_b=\frac{15}{4}\varepsilon N\frac{\Delta\Omega}{4\pi}\frac{\Delta\Omega_T}{4\pi}, \qquad (26)$$

where ε and N are defined in Sec. 2, and $\Delta\Omega/4\rho$ is the relative solid angle within which the L_α photons are recorded. From the kinetic equations describing the stationary populations of the fine-structure levels of o-Ps in the presence of both microwave and laser fields, we can readily determine the increase in the population of the $2\,^3P_2$ level due to the laser radiation, and the corresponding increase in the coincidence signal:

$$i_u=\frac{3}{4}N\frac{\bar{W}}{W_T^{(3)}}\frac{v}{V}\frac{\Delta\Omega}{4\pi}\frac{\Delta\Omega_T}{4\pi}, \qquad (27)$$

where v/V is the ratio of the volume illuminated by the laser radiation to the volume excited by the microwave field. If we substitute $v/V=s/S$ in (27), where s is the cross section of the laser beam and S the cross section of the microwave cavity, and if we recall that the minimum diameter of the laser beam is restricted by the requirement that transit broadening must be small in comparison with the broadening due to the quadratic Doppler effect ($d\gtrsim u/\Delta\nu_q\approx 0.5$ mm), we can use (26), (27), and (25) to determine the laser radiation power P for which the coincidence signal stimulated by the laser field at the resonance maximum is of the order of the background coincidence signal. Assuming, as before, that $\varepsilon = 10^{-4}$ and that the minimum cross sections are $s = 2\times10^{-3}$ cm^2 and $S = 1$ cm^2, [1] we find that $P \approx 500$ W. The pulsed dye laser[17] can, in fact, be used to produce such (and much higher) laser power levels at $\lambda = 4860$ Å.

Let us now determine the detection time when a pulsed laser source is employed. Let τ_p be the length of the laser pulse and T the time between the pulses. For a given signal-to-noise ratio S/N, the minimum detection time is then given by

$$\tau=\left(\frac{S}{N}\right)^2\frac{i_b+i_u\tau_p/T}{i_u^2}\left(\frac{T}{\tau_p}\right)^2. \qquad (28)$$

Assuming that $S/N = 3$, $\Delta\Omega/4\pi = \Delta\Omega_\gamma/4\pi = 0.25$, $N = 10^5$ positronium atoms per second, laser power $P = 5\times10^4$ W, pulse length $\tau_p = 10^{-8}$ sec, [17] repetition frequency $T^{-1} = 1000$ Hz, and the other parameter values used before, we find that $\tau \approx 400$ sec at the resonance maximum. A total of about 880 background coincidences and about 90 coincidences stimulated by the laser radiation will be recorded during this time.

The other possibility of recording the narrow two-photon $1\,^3S_1-2\,^3S_1$ absorption resonance is based on the observation of the 2γ-annihilation decay of the mixed $2\,^3S_1$ state in a magnetic field. Estimates show that the observation time is now greater by roughly an order of magnitude.

4. NARROW RESONANCES IN 2γ-ANNIHILATION RADIATION

In addition to the above methods of Doppler-free laser spectroscopy of positronium, there is considerable independent interest in the possibility of narrow frequency-tunable resonances in 2γ-annihilation radiation, which is specific for the positronium atom. This possibility is based on velocity-selective excitation of positronium atoms through an allowed optical transition by the progressive laser wave, and the subsequent conversion of the excited atoms to states which annihilate into two gamma rays with a high probability.

In actual fact, a progressive laser wave of frequency ω will induce a transition of frequency ω_0 only in the case of atoms with a definite velocity component

$$v_0\approx(\omega-\omega_0)/k \qquad (29)$$

along the direction of the wave vector **k**. If the conversion mechanism ensures that only the excited atoms are transferrred to the state that decays into two gamma rays, then, provided the observations are carried in a direction parallel (or antiparallel) to the direction of propagation of the light wave, one will record a narrow 2γ-annihilation resonance from the selectively excited positronium atoms against the background of the Doppler-broadened annihilation line due to the "non-selected" positronium atoms. The width of the narrow resonance is determined by the width Γ of the optical resonance at the transition frequency ω_0:

$$\Gamma_\gamma=\Gamma\omega_\gamma^0/\omega_0,\quad \Gamma=\Gamma_1\sqrt{1+G}, \qquad (30)$$

and the frequency

$$\omega_\gamma\approx\omega_\gamma^0\pm k_\gamma v_0 \qquad (31)$$

will depend on the laser frequency. When the latter frequency is varied within the limits of the Doppler profile of the optical transition, the narrow 2γ-annihilation resonance will shift within the limits of the Doppler profile of the annihilation radiation.

For the ground and the first excited states of the positronium atom, there are two possible methods of producing narrow 2γ-annihilation resonances. The first involves the direct excitation of o-Ps atoms by a progressive light wave into the $2\,^3P_0$ or $2\,^3P_2$ state which decays into two gamma rays. The second involves the selective excitation of positronium atoms in a magnetic field, for example, through the $1\,^3S_1(m=\pm1)-2\,^3P_1(m=0)$ transition followed by conversion into the $2\,^3P_1(m=\pm1)$ state by the microwave field. As a result of spontaneous L_α decay, the selectively excited and converted atoms are found in the mixed $1\,^3S_1(m=0)$ state which, in a sufficiently strong magnetic field, will decay into two gamma rays with very high probability. We note that the necessity for the microwave field is dictated by the fact that the $1\,^3S_1(m=0)$ and $1\,^3S_1(m=\pm1)$ states have magnetic quantum numbers m_S differing by unity and, since in the optical transition $\Delta m_S = 0$, the atoms excited to the $2\,^3P_1(m=0)$ state cannot undergo radiative transitions to the mixed $1\,^3S_1(m=0)$ state. However, if the microwave field stimulates magnetic transitions ($\Delta m_S = \pm1$) between the $2\,^3P_1(m=0)$ and $2\,^3P_1(m=\pm1)$ states, the L_α decay to the $1\,^3S_1(m=0)$ state is possible from the latter state.

In the discussion below, we shall confine our attention to the first possibility and, to be specific, we will take the level $2\,^3P_0$. The use of the second method encounters considerable difficulties because the $1\,^3S_1$ level is the ground state and its population is greater than that of the $2\,^3P_0$ level by the factor

$$({}^3/_4N\tau_T^{(0)})/({}^3/_4\varepsilon N\tau_{L_\alpha})\approx4.4\cdot10^5 \qquad (32)$$

In fact, under optimum conditions, the fraction $\varkappa$ of atoms that are selectively excited and converted to the $1\,^3S_1(m=0)$ and $2\,^3P_0$ states is practically the same but if, for the $1\,^3S_1(m=0)$ level, the ratio of the intensity of the 2γ-annihilation radiation in the narrow line to the intensity of the annihilation radiation in the broad Doppler line is

$$\left(\frac{i_u}{i_b}\right)_{1S}\approx\frac{3}{4}N\frac{\tau_T^{(0)}}{\tau_S^{(0)}}\varkappa\Big/\frac{3}{4}N\frac{\tau_T^{(0)}}{\tau_S^{(0)}}=\varkappa, \qquad (33)$$

then, for the $2\,^3P_0$ level, this ratio is much greater, i.e.,

$$\left(\frac{i_u}{i_b}\right)_{2P}\approx\frac{3}{4}N\frac{\tau_T^{(0)}}{\tau_T^{(1)}}\varkappa\Big/\frac{3}{4}\varepsilon N\frac{\tau_{L\alpha}}{\tau_T^{(1)}}=\frac{\tau_T^{(0)}}{\tau_{L_\alpha}}\frac{\varkappa}{\varepsilon}\approx4.4\cdot10^5\varkappa. \qquad (34)$$

Figure 4 shows a possible experimental arrangement for producing and detecting the narrow 2γ-annihilation resonance in the case of the $2\,^3P_0$ state. The gamma-ray counters are used in coincidence in order to eliminate a substantial part of the broad Doppler line of annihilation radiation. This system will record both the resonance gamma rays and the 2γ-annihilation radiation from the o-Ps atoms formed in the $2\,^3P_0$ and $2\,^3P_2$ states, and that part of the 3γ-annihilation radiation from o-Ps atoms which contains two gamma rays with energies $mc^2(1-\delta)\leq E \leq mc^2$. The background coincidence signal will, as before, be given by (6) and (7).

The shape of the narrow 2γ-annihilation resonance will be determined by the number of atoms with velocity $\mathbf{v}$ in the interval d^3v that are excited to the $2\,^3P_0$ state:

$$dn_T^{(1)}(\mathbf{v})=(N_T^{(0)}-N_T^{(1)})\frac{\gamma_0}{\gamma_0+\gamma_1}\frac{G}{1+G}\frac{\Gamma^2/4}{(\omega'-\omega_0)^2+\Gamma^2/4}w(\mathbf{v})\,d^3v, \tag{35}$$

where

$$\omega'=\omega(1-\mathbf{n}\mathbf{v}/c)(1-v^2/c^2)^{-1/2}, \tag{36}$$

and $\mathbf{n}=\mathbf{k}/k$. If we use (36) and the change of variables $v,\ \vartheta,\ \varphi\to\omega_\gamma,\ \vartheta,\ \varphi$ in (35), where the frequency of the gamma rays recorded in the direction of $\mathbf{n}_\gamma=\mathbf{k}_\gamma/k_\gamma=\pm\mathbf{n}$ is

$$\omega_\gamma=\omega_\gamma^0(1-v^2/c^2)^{1/2}/(1-\mathbf{v}\mathbf{n}_\gamma/c), \tag{37}$$

and if we integrate with respect to the angular variables, we obtain the following expression for the line shape of the narrow resonance in the 2γ-annihilation radiation recorded in the direction of $\mathbf{n}_\gamma$:

$$\mathcal{I}_{\mathbf{n}_\gamma}(\omega_\gamma)=\int\frac{dn_T^{(1)}(\omega_\gamma,\vartheta,\varphi)}{\tau_T^{(1)}}\sin\vartheta\,d\vartheta\,d\varphi\frac{\Delta\Omega_\gamma}{4\pi}. \tag{38}$$

If the narrow 2γ-annihilation resonance is observed in the direction of $\mathbf{n}_\gamma=\mathbf{n}$, the shape of the resonance line is Lorentzian:

$$\mathcal{I}_{\mathbf{n}}(\omega_\gamma)\sim\frac{\Gamma_\gamma^2/4}{(\omega_\gamma-\omega_\gamma^0\omega/\omega_0)^2+\Gamma_\gamma^2/4}, \tag{39}$$

where Γ_γ is determined by the ratio (30) and the central frequency of the resonance is $\omega_\gamma^0\omega/\omega$ (Fig. 5). The line shape observed in the opposite direction ($\mathbf{n}_\gamma=-\mathbf{n}$) is more complicated. However, in the first approximation in v/c, i.e., to within the linear Doppler effect, it is also described by the Lorentz curve

$$\mathcal{I}_{-\mathbf{n}}(\omega_\gamma)\sim\frac{\Gamma_\gamma^2/4}{[\omega_\gamma-\omega_\gamma^0\omega/(2\omega-\omega_0)]^2+\Gamma_\gamma^2/4}. \tag{40}$$

We note that, in the derivation of (38), we have neglected the natural width $\Gamma_\gamma^{(0)}=1/2\pi\tau_T^{(1)}\approx 1.6$ kHz of the annihilation radiation which, even for $G=0$, is much less than $\Gamma_\gamma\approx 5\times10^{12}$ Hz.

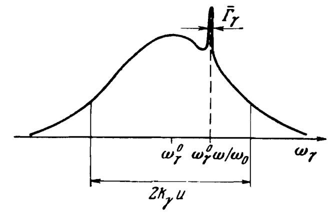

FIG. 5. Narrow resonance in the 2γ-annihilation radiation on the Doppler profile of the annihilation radiation.

It is important to note that the width Γ_γ of the narrow 2γ-annihilation resonance, due to the width of the resonance involving the optical transition, is the lower limit. In a real experiment, the observed width $\bar{\Gamma}_\gamma$ of the resonance is always greater because of the geometric broadening resulting from the detection of the gamma rays in a finite solid angle $\Delta\Omega_\gamma$:

$$\bar{\Gamma}_\gamma=\Gamma_\gamma+\Gamma_{\text{geom}}. \tag{41}$$

For this reason, the limiting narrowing of the 2γ-annihilation line

$$k_\gamma u/\Gamma_\gamma\approx 8.4\cdot10^3(1+G)^{-1/2} \tag{42}$$

is difficult to achieve, and the real narrowing of the line is determined by the required time of detection based on (5) and (28), where, according to (35),

$$i_u=\frac{3}{4}N\frac{\tau_T^{(0)}}{\tau_T^{(1)}}\sqrt{\pi}\frac{\Gamma}{k_\gamma u}\frac{G}{1+G}\frac{\gamma_0}{\gamma_0+\gamma_1}\frac{\Delta\Omega_\gamma}{4\pi}. \tag{43}$$

Suppose, for example, that the required narrowing of the 2γ-annihilation line is $k_\gamma u/\bar{\Gamma}_\gamma\approx 10^2$. Assuming that we use continuous laser radiation with power density $I\approx 3$ W/cm^2 ($G\approx 1$) and that the upper limit for the solid angle is $\Delta\Omega_\gamma\lesssim 8\bar{\Gamma}_\gamma/k_\gamma u\approx 0.08$, we can use (5), (7), and (43) to show that, if the positronium atom concentration is $N=10^5$ sec^{-1}, the time of detection is $\tau\approx 1000$ sec. Finally, the broadening of the narrow resonance up to $\bar{\Gamma}_\gamma$ can always be obtained by increasing the angular divergence of the light wave.

We note in conclusion that narrow 2γ-annihilation lines produced by positronium atoms are of interest in connection with the problem of the gamma laser. As a matter of fact, the amplification of the 2γ-annihilation radiation in a standing γ-wave (without taking into account the quadratic Doppler effect) can be increased by a factor of $\sim k_\gamma u/\Gamma$ at the maximum of the narrow line.

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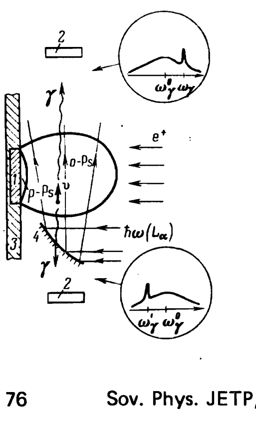

FIG. 4. Possible scheme for the production and detection of the narrow resonance in the 2γ-annihilation radiation from o-Ps atom; 1—target, 2—gamma-ray counters, 3—shield, 4—defocusing mirror.

[1)] The contribution of the $2P$ state to (18) was not taken into account in[16], so that the result $B=\frac{1}{2}A^2\approx 6$ was obtained instead of the correct result $B\approx 243$.

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Scattering of slow electrons by polar molecules

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The scattering of electrons by polar molecules is considered for the energy range in which, on the one hand, the molecule may be considered as immobile, and on the other, the electron wavelength is large in comparison with the radius of short-range forces. The existence of dipole resonances is discussed. The results of the theory are compared with the experimental data and calculations are carried out by the close-coupling method.

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1. INTRODUCTION

The fundamental contribution to the scattering of slow electrons by polar molecules is made by the long-range electron-dipole interaction. Therefore, only this part of the interaction between electron and molecule has been taken into account in a number of researches on the calculation of the scattering cross section. Scattering has been considered by an immobile[1] and by a rotating[2] dipole in the Born approximation. In the first case, the total cross section diverges, owing to the long-range character of the interaction,[3] while in the second case, if the molecule is regarded as a rigid rotator, we get[1)]

$$\sigma(j\to j\pm1)=\frac{8\pi}{3k_i^2}D^2\frac{j_>}{2j+1}\ln\left|\frac{k_i+k_f}{k_i-k_f}\right|, \quad (1)$$

where j is the rotational quantum number of the rotator, $j_>=\max(j, j\pm1)$, D is the dipole moment, while k_i and k_f are the initial and final wave numbers of the electron and are connected by the relation

$$k_i^2+j(j+1)/I=k_f^2+(j\pm1)(j\pm1+1)/I, \quad (2)$$

where I is the moment of inertia of the rotator.

Mittleman and von Holdt[4] have given an exact solution of the problem of scattering by an immobile point dipole. Because of the singularity of the potential $D\cos\theta/r^2$ at zero, such an approach is limited to values of $D<D_{cr}=0.639$. At $D\geqslant D_{cr}$, collapse to the center takes place. Therefore a more realistic approach is the consideration of scattering by a finite dipole. This problem has been solved both numerically[5] and analytically.[6]

Simultaneous account of strong interaction and rotation of the molecule has been carried out by the method of strong coupling,[7] used in the consideration of scattering by a whole series of molecules.

In all these researches, the short-range part of the interaction has not been computed from first principles, but introduced in the form of a model potential, which introduces some ambiguity in the interpretation of the