cm/sec. The uncertainty given for v_r is due to an allowed $\pm 10°C$ uncertainty in the cell temperature as measured by a mercury thermometer placed in the oven containing the optical pumping cell.

The data of this experiment yields a spin-exchange cross section for Rb^{85}-Rb^{87} collisions of $(1.70\pm0.21)\times10^{-14}$ cm^2. The early work of Franken, Sands, and Hobart[5] and Novick and Peters[6] yielded cross sections of 5×10^{-14} cm^2 for Na-K collisions and 2×10^{-14} cm^2 for Na-Rb^{85} collisions. These results were considered to be reliable within a factor of 3 of the quoted values. The uncertainty was primarily due to unreliable density estimates. With the use in the present work of the interferometric technique of density measurement, estimated to be reliable to $\pm 11\%$, this type of uncertainty was largely overcome. The total error quoted for the cross section is closely related to a 70% confidence interval. The result presented is in reasonable agreement with measurements of Rb^{85}-Rb^{85} and Rb^{87}-Rb^{87} spin-exchange collision cross sections performed by Moos and Sands[18] in this laboratory using an electron paramagnetic resonance technique.

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[18] T. Stark and R. H. Sands have kindly informed us by private communication of recent results for both isotopes of Rb based on the Ph.D. thesis of H. W. Moos at the University of Michigan, 1961 (to be published).

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Interaction of Optical and Infrared Radiation with Metastable Hydrogen Atoms

WOLFGANG ZERNIK
Plasma and Space Applied Physics, Radio Corporation of America, Princeton, New Jersey

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This paper is an extension of a previous one and discusses the theory of the quenching of the metastable $2S$ state of atomic hydrogen by means of optical radiation, for example by the light from a ruby laser. The case discussed is that for which the incident intensity is sufficiently weak for the usual quantum-electrodynamical perturbation theory to be valid. A procedure developed by Schwartz and others is used to carry out the sum over intermediate states without explicit enumeration. The results are given for a range of incident wavelengths from 5000 Å to 50 μ. For unpolarized light from a ruby laser (6934 Å), the total cross section for quenching is found to be $\sigma_Q=1.27\times10^{-22}$ cm^2. The cross section for coherent scattering has also been calculated for the same range of wavelengths; for ruby laser light, the total cross section for scattering is found to be $\sigma_S=1.03\times10^{-23}$ cm^2.

1. INTRODUCTION

IN a previous paper,[1] hereinafter referred to as I, an approximate calculation was reported for the quenching of the atomic hydrogen $2S$ state by means of the light from a ruby laser. It was concluded that, (a) the process can be analyzed by means of the usual quantum-electrodynamical perturbation theory[2] provided that the peak electric-field strength in the laser beam is less than about 10^7 V/cm, and that if this is the case, then, (b) the process consists mainly of virtual excitation to the $3P$ state followed by spontaneous decay to the $1S$ ground state.

In this paper the treatment is restricted to the weak-field case but all possible intermediate states, including those in the continuum, are taken into account.

[1] W. Zernik, Phys. Rev. **132**, 320 (1963).
[2] See, for example, W. Heitler, *The Quantum Theory of Radiation* (Clarendon Press, Oxford, 1954), 3rd ed.

In addition, numerical results for the quenching cross section are given for a range of incident wavelengths from 5000 Å to 50 μ, essentially covering the range of currently available laser frequencies. The maximum intensity of the incident radiation for which the results are valid is a function of the frequency, as explained in I. In particular, the theory breaks down completely in the neighborhood of the Balmer frequencies.

2. PERTURBATION THEORY RESULTS

As indicated in I, second-order perturbation theory yields a cross section for quenching given by:

$$\frac{d\sigma}{d\Omega}=r_0^2\left\{k_1^3k_0\left|\sum_i\frac{(\boldsymbol{\varepsilon}_0\cdot\mathbf{r}_{i0})(\boldsymbol{\varepsilon}_1\cdot\mathbf{r}_{fi})}{E_0-E_i+k_0}+\frac{(\boldsymbol{\varepsilon}_1\cdot\mathbf{r}_{i0})(\boldsymbol{\varepsilon}_0\cdot\mathbf{r}_{fi})}{E_0-E_i-k_1}\right|^2 +k_2^3k_0\left|\sum_i\frac{(\boldsymbol{\varepsilon}_0\cdot\mathbf{r}_{i0})(\boldsymbol{\varepsilon}_2\cdot\mathbf{r}_{fi})}{E_0-E_i-k_0}+\frac{(\boldsymbol{\varepsilon}_2\cdot\mathbf{r}_{i0})(\boldsymbol{\varepsilon}_0\cdot\mathbf{r}_{fi})}{E_0-E_i-k_2}\right|^2\right\}. \quad (1)$$

In this equation, r_0 is the classical electron radius in cm and all other quantities are in atomic units, i.e., energies in units of $(me^4/\hbar^2)$ and dipole matrix elements in units of $(\hbar^2/me^2)$. Subscripts 0, i, and f refer to initial intermediate, and final states, respectively. The $\mathbf{r}_{i0}$ are dipole matrix elements, the $\boldsymbol{\varepsilon}_0$ are unit polarization vectors, and E denotes the energy of an atomic state. The wave number of the incident photon, in energy units, is denoted by k_0. The wave numbers of the two possible emitted photons k_1 and k_2, are given by

$$k_1=E_0-E_f+k_0, \tag{2}$$

$$k_2=E_0-E_f-k_0. \tag{3}$$

The four terms in (1) may be regarded as representing the following paths by means of which the process occurs: (a) k_0 is absorbed, then k_1 is emitted, (b) k_1 is emitted, then k_0 is absorbed, (c) another k_0 is emitted, then k_2 is emitted, (d) k_2 is emitted, then another k_0 is emitted.

The summation over intermediate states i, in Eq. (1), includes all those states allowed by the dipole selection rules, i.e., $2P$, $3P$, $4P$, $\cdots$, etc.

It was indicated in I that the angular factors in (1) may be taken out and the result written in the following form

$$\frac{d\sigma}{d\Omega}=2Q(k_0,k_1)[|a_\perp^{(1)}|^2+|a_{\parallel}^{(1)}|^2]+2Q(k_0,k_2)[|a_\perp^{(2)}|^2+|a_{\parallel}^{(2)}|^2]\ \text{cm}^2/\text{sr}. \tag{4}$$

In (4), $a_\perp$ and $a_\parallel$ are the inelastic scattering amplitudes for final-state polarizations perpendicular and parallel to the scattering plane, respectively. They are determined by

$$a_\perp^{(1)}=\epsilon_{0\perp}, \tag{5}$$

$$a_\parallel^{(1)}=\epsilon_{0\parallel}\cos\theta_1; \tag{6}$$

here θ_1 is the angle between $\mathbf{k}_0$ and $\mathbf{k}_1$, and $\epsilon_{0\perp}$, $\epsilon_{0\parallel}$ are the components of the incident unit polarization vector perpendicular and parallel to the scattering plane, with

$$\epsilon_{0\perp}^2+\epsilon_{0\parallel}^2=1. \tag{7}$$

Therefore, if the incident light is unpolarized and the polarization of the inelastically scattered light is not measured, the cross section for quenching is

$$\frac{d\sigma}{d\Omega}=Q(k_0,k_1)[1+\cos^2\theta_1]+Q(k_0,k_2)[1+\cos^2\theta_2]\ \text{cm}^2/\text{sr}. \tag{8}$$

The quantities $Q(k_0,k_1)$ and $Q(k_0,k_2)$ in Eq. (4), may be expressed as

$$Q(k_0,k_1)=(1/18)r_0^2\{k_1^3k_0|P(k_0)+P(-k_1)|^2\}, \tag{9}$$

$$Q(k_0,k_2)=(1/18)r_0^2\{k_2^3k_0|P(-k_0)+P(-k_2)|^2\}, \tag{10}$$

where

$$P(k)=\sum_{i=2P}^{\infty}P_i(k), \tag{11}$$

and

$$P_i(k)=\frac{r_{0i}r_{if}}{E_0-E_i+k}. \tag{12}$$

The special case of light from a ruby laser (6934 Å) will now be considered and estimates made of the contributions of several possible intermediate states to the cross section for quenching. These calculations are useful for understanding the physics involved in the process and as rough check on the exact but somewhat indirect calculations to be described in the next section.

For incident ruby light, $k_0=0.0657$ $(me^4/\hbar^2)$ which is just slightly less than $E_{3P}-E_{2S}=0.0695$ $(me^4/\hbar^2)$. Therefore, the largest of the $P_i(k)$ is $P_{3P}(k_0)$. In Table I, values of the $P_i(k)$ for $i\equiv 2P$, $3P$, and $4P$ are given. The $P(k)$ are obtained by the method described in the next section.

One notes that the transition via the $2P$ state contributes a not insignificant amount to the cross section and that this transition does not saturate (as does that via the $3P$ state) when the field strength in the incident beam approaches about 10^7 V/cm.

TABLE I. Values of $P_i(k)$ for incident ruby laser light. $P_i(k)=(r_{2S,i}r_{i,1S})/(E_{2S}-E_i+k)$. The $r_{2S,i}$ are dipole matrix elements between atomic states having energies E_{2S}, E_i; k_0 is the energy of the incident photons; $k_1=E_{2S}-E_{1S}+k_0$; $k_2=E_{2S}-E_{1S}-k_0$. Matrix elements and energies are in units of $\hbar^2/me^2$, $me^4/\hbar^2$, respectively. $P(k)=\sum_{i=2P}^{\infty}P_i(k)$.

i	$+k_0$	$-k_1$	$-k_0$	$-k_2$
$2P$	−102.2	+15.2	+102.2	+21.7
$3P$	−412.9	−3.1	−11.6	−4.1
$4P$	−13.9	−0.7	−2.4	−1.0
$P(k)$	−558.8	8.65	80.72	13.06

3. IMPLICIT SUMMATION OVER INTERMEDIATE STATES

In this section, the sum defined by Eqs. (11) and (12) will be evaluated by means of a technique similar to that introduced by Schwartz and Tieman[3,4] and utilized by Mittleman and Wolf[5] in their calculation of the coherent scattering of photons by atomic hydrogen in the ground state.

Denoting the normalized radial functions[6] for hydrogen by $R_{nl}(r)$, one defines the function

$$U(r,k)=\sum_{n=2}^{\infty}\frac{rR_{n1}(r)\int_0^\infty R_{n1}(r')R_{20}(r')r'^3dr'}{E_2-E_n+k}. \tag{13}$$

[3] C. Schwartz, Ann. Phys. (N. Y.) 6, 156 (1959).
[4] C. Schwartz and T. J. Tieman, Ann. Phys. (N. Y.) 6, 178 (1959).
[5] M. H. Mittleman and F. A. Wolf, Phys. Rev. 128, 2686 (1962).
[6] See, for example, H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One and Two Electron Atoms* (Academic Press Inc., New York, 1957), Sec. 3.

Using (11) and (13) one obtains

$$P(k)=\int_0^\infty R_{10}(r)U(r,k)r^2dr. \tag{14}$$

The functions $rR_{n1}(r)$ satisfy the equation

$$\frac{1}{2}\frac{d^2}{dr^2}[rR_{n1}(r)]+\left[\frac{1}{r}-\frac{1}{r^2}\right]rR_{n1}(r)=-E_n rR_{n1}(r), \tag{15}$$

and the orthonormality and closure conditions:

$$\int_0^\infty r^2R_{n1}(r)R_{m1}(r)dr=\delta_{nm}, \tag{16}$$

$$\sum_{n=2}^{\infty} rR_{n1}(r)r'R_{n1}(r')=\delta(r-r'). \tag{17}$$

Consequently, the function $U(r,k)$ satisfies the equation

$$\left[E_2+k+\frac{1}{2}\frac{d^2}{dr^2}+\frac{1}{r}-\frac{1}{r^2}\right]U(r,k)=R_{20}(r)r^2. \tag{18}$$

One now introduces the Laplace transform of $U(r,k)$,

$$S(p,k)=\int_0^\infty U(r,k)e^{-pr}dr, \tag{19}$$

and notes the relations

$$\int_0^\infty r^te^{-pr}U(r)dr=\left[\left(-\frac{d}{dp}\right)^t S(p)\right], \tag{20}$$

and

$$\int_0^\infty r^2e^{-pr}\frac{d^2U}{dr^2}dr=\frac{d^2}{dp^2}[p^2S(p)]. \tag{21}$$

Using (20) and (21) in (18) one obtains

$$(-\tfrac{1}{8}+k+\tfrac{1}{2}p^2)\frac{d^2S}{dp^2}+(2p-1)\frac{dS}{dp}$$
$$=2^{-1/2}[24(p+\tfrac{1}{2})^{-5}-60(p+\tfrac{1}{2})^{-6}]. \tag{22}$$

Eq. (22) is a first-order differential equation which may readily be solved numerically. However, it is important to specify the boundary conditions correctly and this is done as follows.

From the definition of $S(p,k)$ as the Laplace transform of a function that goes to zero exponentially as $r\rightarrow\infty$, it follows that $S(p,k)$ and all its derivatives must be finite for $\mathrm{Re}\,p>0$. Hence, one may put $\frac{1}{2}p^2=(+\frac{1}{8}-k)$ in Eq. (22) and obtain dS/dp at this point, for all $k<\frac{1}{8}$.

One may now integrate the equation numerically to $p=1$, obtaining dS/dp and, hence, d^2S/dp^2 at this point.

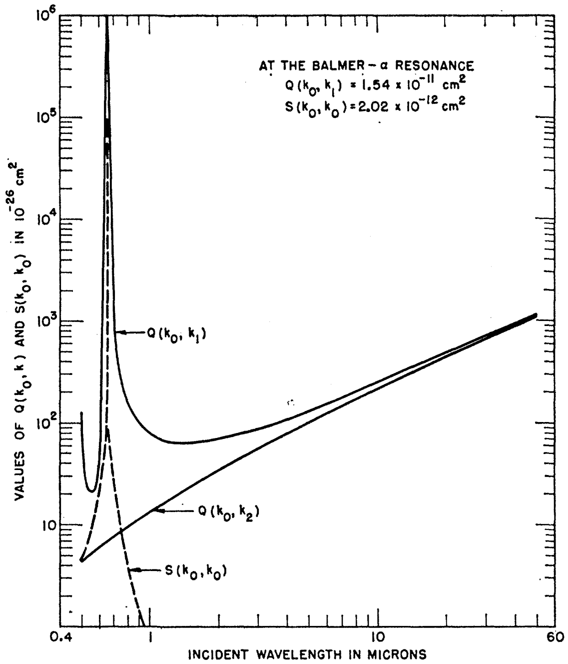

FIG. 1. Graphs of the differential cross-section coefficients $Q(k_0,k_1)$, $Q(k_0,k_2)$, and $S(k_0,k_0)$ versus the wavelength of the incident light.

Using (20) and (14) one finds that

$$P(k)=2\left[\frac{d^2S(p,k)}{dp^2}\right]_{p=1}. \tag{23}$$

This expression has been evaluated numerically and the resulting values of $P(k)$ required for incident ruby laser light are given in Table I. The resulting values of $Q(k_0,k_1)$ and $Q(k_0,k_2)$ are

$$Q(k_0,k_1)=7.539\times10^{-24}\ \mathrm{cm^2/sr}, \tag{24}$$

$$Q(k_0,k_2)=7.542\times10^{-26}\ \mathrm{cm^2/sr}. \tag{25}$$

The differential cross sections for quenching are given by Eqs. (4) or (8). The total cross section for quenching by ruby laser (6934 Å) light works out to be

$$\sigma_Q=1.27\times10^{-22}\ \mathrm{cm^2}. \tag{26}$$

Figure 1 shows graphs of $Q(k_0,k_1)$ and $Q(k_0,k_2)$ versus the wavelength of the incident beam,[7] from 5000 Å to 50 μ. One notes the expected resonance in the neighborhood of the Balmer-α line (6563 Å). These graphs enable one to calculate the results of any possible quenching experiment carried out with currently available lasers, provided only that the criterion for a "weak-field" treatment, which is given in **I**, is satisfied.[8]

[7] The peak of the Balmer-α resonance was calculated by means of the strong field theory as in **I**.

[8] One notes that the "strong-field" theory described in **I** takes only one intermediate state into account, so that it can not be used to obtain *exact* results in the weak-field limit except at a resonance peak.

Table II. Values of $Q(k_0, k_1)$, $Q(k_0, k_2)$, and $S(k_0, k_0)$ for incident wavelengths from 5000 Å to 50 μ. The energy of the incident photons is k_0, corresponding to a wavelength λ_0; $k_1=E_{2S}-E_{1S}+k_0$, $k_2=E_{2S}-E_{1S}-k_0$. If the incident light is unpolarized, and the polarization of the outgoing light is not measured, then the differential cross sections for quenching with emission of k_1 or k_2, respectively, are $(d\sigma/d\Omega)_1=Q(k_0,k_1)[1+\cos^2\theta_1]$, $(d\sigma/d\Omega)_2=Q(k_0,k_2)\times[1+\cos^2\theta_2]$, where θ_1 and θ_2 are the angles between k_0 and k_1 or k_2, respectively. The differential cross section for scattering of unpolarized light is given by $(d\sigma/d\Omega)_S=S(k_0,k_0)[1+\cos^2\theta_S]$, where θ_S is the scattering angle.

λ_0 (microns)	$Q(k_0,k_1)$ in 10^{-26} cm^2	$Q(k_0,k_2)$ in 10^{-26} cm^2	$S(k_0,k_0)$ in 10^{-26} cm^2
0.5	1.296×10^2	4.458	4.643
0.6	4.277×10	6.002	2.086×10
0.6550	4.816×10^5	6.899	6.576×10^4
0.6563	$1.54\ \times10^{15}$		$2.02\ \times10^{14}$
0.6570	7.670×10^5	6.932	1.015×10^5
0.6934	7.539×10^2	7.542	6.122×10
0.7	5.909×10^2	7.654	4.444×10
0.8	1.409×10^2	9.392	4.190
0.9	9.334×10	1.120×10	1.386
1	7.751×10	1.306×10	6.468×10^{-1}
2	7.150×10	3.347×10	1.869×10^{-2}
3	9.060×10	5.525×10	3.306×10^{-3}
4	1.120×10^2	7.753×10	1.008×10^{-3}
5	1.342×10^2	1.000×10^2	4.060×10^{-4}
6	1.566×10^2	1.227×10^2	1.940×10^{-4}
7	1.792×10^2	1.454×10^2	1.042×10^{-4}
8	2.019×10^2	1.682×10^2	6.084×10^{-5}
9	2.246×10^2	1.910×10^2	3.790×10^{-5}
10	2.474×10^2	2.138×10^2	2.482×10^{-5}
20	4.763×10^2	4.428×10^2	1.542×10^{-6}
30	7.055×10^2	6.721×10^2	3.045×10^{-7}
40	9.349×10^2	9.014×10^2	9.640×10^{-8}
50	1.164×10^3	1.131×10^3	3.940×10^{-8}

Since it will not be possible for the reader to read accurate numbers off the graph, the results are also given in tabular form in Table II.[9,10]

4. COHERENT SCATTERING

The previous calculations also enable one to calculate the coherent scattering cross section with little additional work.

Perturbation theory yields a result which may be written down by analogy with Eq. (4),

$$d\sigma/d\Omega = 2S(k_0,k_0)[|a_\perp^{(S)}|^2+|a_\parallel^{(S)}|^2]\ \text{cm}^2/\text{sr}, \tag{27}$$

where $a_\perp^{(S)}$ and $a_\parallel^{(S)}$ are the elastic-scattering amplitudes for final-state polarizations perpendicular and parallel to the scattering plane. If the incident light is unpolarized and the polarization of the scattered light is not measured, Eq. (27) becomes:

$$d\sigma/d\Omega = S(k_0,k_0)[1+\cos^2\theta_S], \tag{28}$$

where θ_S is the scattering angle.

The coefficient $S(k_0,k_0)$ is given by

$$S(k_0,k_0)=(1/18)r_0^2\{k_0^4|F(k_0)+F(-k_0)|^2\}, \tag{29}$$

where

$$F(k)=\sum_{i=2P}^{\infty} F_i(k), \tag{30}$$

and

$$F_i(k)=\frac{r_{0i}r_{i0}}{E_0-E_i+k}. \tag{31}$$

Now using Eq. (13), one finds that $F(k)$ is given by

$$F(k)=\int_0^\infty R_{20}(r)U(r,k)r^2dr, \tag{32}$$

which, using Eq. (20), may be written as

$$F(k)=2^{-1/2}\left[\left(\frac{d^2S}{dp^2}\right)_{p=1/2}+\frac{1}{2}\left(\frac{d^3S}{dp^3}\right)_{p=1/2}\right]. \tag{33}$$

Using Eq. (22), one finds finally

$$F(k)=\frac{1}{k}\left[42+\frac{9}{2k}-2^{-1/2}\left(\frac{dS}{dp}\right)_{p=1/2}\right]. \tag{34}$$

This quantity may be evaluated by solving Eq. (22) numerically as explained in Sec. 3. A rough check on the results can be obtained by calculations analogous to those summarized in Table I.

For ruby laser light, one finds

$$S(k_0,k_0)=6.122\times10^{-25}\ \text{cm}^2/\text{sr}, \tag{35}$$

and the total coherent scattering cross section is

$$\sigma_S=1.03\times10^{-23}\ \text{cm}^2. \tag{36}$$

A graph of $S(k_0,k_0)$ versus the wavelength of the incident beam is shown on Fig. 1.

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[9] It has been brought to the author's attention by J. P. Wittke of RCA Laboratories that the wavelength used for ruby laser light in this work, i.e., 6934 Å, is actually that appropriate to liquid-nitrogen temperatures. At room temperature, the wavelength is 6943 Å.

[10] After this work was completed, results of some similar calculations were published by I. D. Abella, M. Lipeles, and N. Tolk [Bull. Am. Phys. Soc. **8**, 476 (1963)]. These results are currently being revised (personal communication from I. D. Abella). It would appear that a quenching experiment with ruby laser light might be more easily done with He^+ metastables rather than H [M. Lipeles, L. Gampel, and R. Novick, Bull. Am. Phys. Soc. **7**, 69 (1962)]. The cross sections for He^+ may be obtained from those for H by simple scaling, i.e., by calculating the H results for a wavelength of four times the ruby value. In the notation of Table II, the results are: $Q(k_0,k_1)=8.600\times10^{-25}$ cm^2; $Q(k_0,k_2)=5.032\times10^{-25}$ cm^2; $S(k_0,k_0)=4.576\times10^{-29}$ cm^2.