Investigation of transitions between orthohydrogen and parahydrogen for detecting parity violation in the weak interaction

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Theoretical estimates are made of the probabilities of electron para–ortho transitions in hydrogen molecules, which are found to be $\sim 10^{20}$ times less probable than the corresponding electron transitions without reorientation of the nuclear spins. This strong forbiddenness for para–ortho transitions, and also the proximity of states of opposite parity make it possible to use hydrogen to detect the weak interaction. In this case, the weak interaction of the vector current of the electrons with the axial current of the nucleons will be manifested. It is suggested that an experimental arrangement may be used in which hydrogen is cyclically pumped from one cuvette to another. Para–ortho transitions are induced in the main cuvette by circularly polarized laser radiation. In the second cuvette, the increase in the number of orthohydrogen molecules is determined on the basis of the fluorescence intensity for the allowed $E1$ transition. The relationship between the number of para and ortho states of the hydrogen molecule can then be used as a memory for accumulating information on the magnitude of the weak interaction. If para–ortho transitions are alternately induced by left- and right-handed polarized laser radiation, the fluorescence intensity should differ by $\sim 10^{-3}\%$ due to nonconservation of parity by the neutral current.

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Effects of nonconservation of parity in the weak interaction of electrons with nucleons in a nucleus have been widely discussed.[1,2] In an investigation of optical transitions in heavy atoms,[3] one can determine only one weak constant, which characterizes the interaction of the axial current of the electrons with the vector current of the protons. There is therefore great interest in other possible experiments. In the present paper, we discuss an experiment on stimulated para–ortho transitions in hydrogen, which leads to a manifestation of the second constant of the weak current characterizing the interaction of the vector current of the electrons with the axial current of the nucleons.

Molecular hydrogen consists of a mixture of two modifications: the orthomodification with parallel spins of the protons and the paramodification with antiparallel spins of the protons. Because of the difference in parity of the nuclear spin wave functions, the two modifications differ by the symmetry of the rotational function under inversion of the coordinates of the four particles at the center of the molecule. Optical transitions between the para and ortho modifications are forbidden by the nuclear spin. In the ${}^1\Sigma_g^+$ ground state and in the first excited electron state $B^1\Sigma_u^+$ of the hydrogen molecule (which we shall consider), the electron spins are antiparallel. Therefore, only magnetic interactions of the spins of the nuclei with the electron orbital angular momenta can effectively make possible para–ortho transitions.

In the proposed experiment it is suggested that resonance laser radiation should be used to induce a sufficient number of molecules of parahydrogen to go over to the excited orthostate. As a result of intense allowed $E1$ transitions [their probability is $W(E1) \sim 10^7$–10^9 sec^{-1}], these molecules will then go over to the orthosublevels of the ground electron state. Decay of the orthostates back to the parastates will occur exclusively through collisions [the probability of these transitions by spontaneous emission is $\sim 10^{-20}$ sec^{-1} (Refs. 4 and 5)]. According to the estimates of Ref. 4, the time of establishment of equilibrium in the ortho–para system under normal conditions and without allowance for the nonideal nature of the walls is ~3 years. One can therefore assume that for reasonable experimental periods hydrogen will continuously accumulate in the orthostate. Since the number of molecules in the new state is much less than the number of molecules remaining the parastate, it is necessary to ensure purity of the original parahydrogen. Namely, the number of remaining molecules of orthohydrogen in the original mixture must at the least not exceed the number of molecules that make the transition from the para to the orthostate as a result of the experiment. These conditions are certainly satisfied at 6 °K, which corresponds to a hydrogen vapor pressure $\sim 10^{-3}$ mm Hg. Therefore, it is sensible to restrict the treatment to transitions from the ground rotational state of parahydrogen $K=J=0$, where K is the rotational quantum number and J is the total angular momentum of the molecule. We shall consider transitions to the $B^1\Sigma_u^+$ state. Obviously, transitions to the levels $K=0$, $J=1$ and $K=2$, $J=1$ are possible. We shall estimate the probabilities of the $M1$ transitions.

The main contribution to the mixing of the ortho and para states will be made by the operator that takes into account the interaction of the magnetic moment of the nucleus with the magnetic field induced by the electrons at the nuclei:

$$\mathbf{H}_M = \mu_N \frac{e}{mc} \sum_{k,l} \frac{\mu_l}{\mu_N} \frac{[\mathbf{r}_{kl}\mathbf{p}_k]}{\Gamma_{kl}^3} = \mu_N \frac{e\hbar}{mc} \tilde{\mathbf{H}}_M, \tag{1}$$

where the subscripts k and l label the electrons and the nuclei, respectively, and μ_N is the nuclear magneton.

The probability of a $M1$ transition is proportional to

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the square of the matrix element of the magnetic moment of the molecule between the considered states. Going over to a rotating coordinate system and making simple transformations, we find that the required matrix element is proportional to the expression

$$A=\frac{e^2\hbar^2}{2m^2c^2}\mu_N(\langle\psi(^1\Sigma_u{}^+)|\tilde{\mathbf{H}}_L|\psi_{Mu}{}^{\Sigma_g}\rangle+\langle\psi_{Mg}{}^{\Sigma_u}|\tilde{\mathbf{H}}_L|\psi(^1\Sigma_g{}^+)\rangle), \qquad (2)$$

where $\psi(^1\Sigma_{gu}{}^+)$ are the wave functions of the ground state and the first excited state, and $\psi_{Mgu}^{\Sigma g,u}$ are the first-order corrections to these functions (the indices g and u indicate the symmetry of the wave functions under inversion of the electron coordinates). The correction functions $\psi_{Mgu}^{\Sigma g,u}$ are determined by solution of an inhomogeneous Schrödinger equation of the type (8) from our paper Ref. 6 when the operator $\tilde{\mathbf{H}}_M$ and the wave functions $\psi(^1\Sigma_g^+)$ and $\psi(^1\Sigma_u^+)$ are used. The operator $\tilde{\mathbf{H}}_L$ has the form

$$\mathbf{H}_L=\frac{e}{2mc}\sum_k[\mathbf{r}_k\mathbf{p}_k]\equiv\frac{e\hbar}{2mc}\tilde{\mathbf{H}}_L. \qquad (3)$$

The matrix elements A_1 and A_2 in the expression (2), $A=A_1+A_2$, are analogous to the corresponding matrix elements in the expression for paramagnetic screening of the hydrogen molecule[7]:

$$\sigma_p\sim\frac{e^2\hbar^2}{m^2c^2}\langle\psi(^1\Sigma_g{}^+)|\tilde{\mathbf{H}}_L|\psi_{Mg}{}^{\Sigma_g}\rangle. \qquad (4)$$

In the calculation of the nonadiabatic corrections to the screening,[6] we had to construct the functions ψ^{Mu} and calculate the matrix elements of the operator $\tilde{\mathbf{H}}_L$ between these functions and basis functions antisymmetric under inversion of the electron coordinates. The ratio of these matrix elements to the matrix elements of the operator $\tilde{\mathbf{H}}_L$ between the functions $\psi(^1\Sigma_u^+)$ and the symmetric basis functions is ~20. The wave function can be constructed as a linear combination of antisymmetric basis functions with coefficients ~1. Using this circumstance, and also $\sigma_p=0.4\times10^{-6}$, we obtain from our calculations of the paramegnetic screening of the protons of hydrogen an estimate for the expression (2):

$$A\approx2A_1\approx20\sigma_p\mu_N\approx5\cdot10^{-29}\ \text{erg/G}.$$

Then for the required probability, we have

$$W(M1)\approx A^2\omega^3/\hbar c^3\approx10^{-12}\ \text{sec}^{-1}.$$

Here, $\omega/c\approx2\pi\times10^5$ cm^{-1}.

This estimate of $W(M1)$ for the forbidden para–ortho transition is preliminary and evidently holds only in order of magnitude. However, as will be shown below, an estimate of the viability of the proposed experiment does not depend on the probability of this transition in a wide range.

We estimate the admixture of states of opposite parity to the considered levels due to the weak interaction. The potential of the weak interaction of an electron with a nucleon, obtained from the most general considerations is given, for example, in Ref. 1. Since we consider states with antiparallel electron spins, and a para–ortho transition is accompanied by reorientation of the nuclear spin, only the second term of this potential, which contains the spin of the nucleon of the nucleus, will be important for us. We rewrite it in the convenient form

$$V_w(e,n)=Vm\varkappa\sigma_n\left\{\left[\frac{\mathbf{p}_e}{m},\delta(\mathbf{r}_e-\mathbf{r}_n)\right]_+-\left[\frac{\mathbf{p}_n}{M},\delta(\mathbf{r}_e-\mathbf{r}_n)\right]_+\right\}, \qquad (5)$$

where $V=G\hbar^3/2^{3/2}c^2m$; G is the Fermi constant; σ_n, $\mathbf{p}_n$, M, and $\mathbf{r}_n$ are the spin matrix, momentum, mass, and coordinate of the electron; and $\varkappa$ is the constant of the interaction of the vector current of the electron with the axial current of the nucleon. The potential for the molecule will be equal to the sum of such potentials over all nucleons and electrons of the molecule:

$$V_w=\sum_{e,n}V_w(e,n).$$

The operator V_w mixes levels of opposite parity for the same electron term. At the same time, the expectation value of the anticommutator, which contains the electron momentum, is zero. Therefore, in the considered matrix element only the second term in the expression (5) will be important.

The operator V_w also mixes vibration–rotation sublevels of opposite parity for different electron states. At the same time, the matrix element of the first term in (5), which depends on the electron momentum, is nonzero, and one can therefore expect that the matrix element of the operator V_w is M/m times greater than the matrix element between sublevels of the same electron state. For the ground state, this increase will be compensated by the approximately same increase of the energy denominator. Much more effective is the mixing for the upper $B^1\Sigma_u^+$ state, since the vibration–rotation sublevels of this state and the mixed sublevels of opposite parity of the $C^1\pi_u$ state are fairly close to each other. It is well known that it is precisely this proximity that is responsible for the appreciable Λ doubling for the $C^1\pi_u$ state.[8,9]

The largest parity violation effects are attained for the 14th vibrational level of the $B^1\Sigma_u^+$ state ($K=0$). The nearest vibrational level of the $C^1\pi_u$ state ($v=3$, $K=1$) is separated from it by less than 10 cm^{-1}. Therefore, the Λ doubling for the third vibrational level of the $C^1\pi_u$ state also appreciably exceeds the Λ doubling for the other vibrational states with the same rotational quantum number.[8,9] The coefficient of the mixing of the levels is inversely proportional to ΔE, the energy difference between the levels of opposite parity. The data on the value of ΔE between these levels are contradictory: $\Delta E=1.7$ cm^{-1} (Ref. 10) and $\Delta E=4.6$ cm^{-1} (Ref. 11). However, the value of the Λ doubling E_Λ for the neighboring $C^1\pi_u$ level does not agree in either Ref. 10 or Ref. 11 with the correct value E_Λ measured in the recent investigation of Takezava and confirmed by the calculation of Ref. 9. If we combine the data of this paper with the data of Ref. 11, we obtain $\Delta E=7.7$ cm^{-1}. We shall use this value to estimate the coefficient of mixing. In fact, as will be shown below, the estimate of the minimal power of the laser needed to carry out the proposed experiment in realistic times (i.e., the viability of this experiment) does not depend on the value of ΔE

To calculate the matrix element of the operator V_w, it is necessary to take into account the strong perturbation of the level $C^1\pi_u$, $v=3$, $J=K=1$ by the neighboring level $B^1\Sigma_u^+$, $v=14$, $J=K=1$. The interaction of these levels is described by the operator

$$H_{\Sigma\pi}=-\frac{1}{2\mu_0 R^2}(L_-K_+ + L_+K_-), \tag{6}$$

where μ_0 is the reduced mass of the molecule, and $L_\pm$ and $K_\pm$ are spherical components of the operator of the electron orbital angular momentum and the operator of the rotational angular momentum of the molecule. The matrix elements of the operator (6) are calculated in Refs. 8 and 9. In particular, $H_{14,3}=-12.9$ cm^{-1}. Thus, the wave function of the state $C^1\pi_u^+$, $v=3$, $J=K=1$ is a superposition of the unperturbed (adiabatic) wave function of this state and the unperturbed wave function of the state $B^1\Sigma_u^+$, $v=14$, $J=K=1$.

Let E_Σ^0 and E_π^0 be the energies of these levels in the adiabatic approximation. It is known that $E_\Sigma^0-E_\pi^0=14.6$ cm^{-1} (Ref. 8). Therefore $H_{14,3}\approx(E_\Sigma^0-E_\pi^0)$. Thus, the weight of the admixed state $B^1\Sigma_u^+$ in the wave function of the considered state $C^1\pi_u^+$ is not small in the given case. Using the experimental value of the Λ doubling and the calculated value of $H_{14,3}$, we can determine the weight of the unperturbed functions describing the contribution of the pure states $B^1\Sigma_u^+$ and $C^1\pi_u^+$ to the wave function of the considered level: $C_\Sigma=0.50$, $C_\pi=0.87$. We calculate the required matrix element:

$$X=\langle B^1\Sigma_u^+,\ v_\Sigma=14,\ J=1,\ M,\ K=0,\ I=1|V_w|C^1\pi_u^+,\ v_\pi=3,\ J=1,\ M,\ K=1,\ I=0\rangle.$$

Since $K\geqslant\Lambda$, the $^1\Sigma_u^+$ state with rotational angular momentum $K=0$ is not perturbed, since the operator $H_{\Sigma\pi}$ does not depend on the nuclear spin and mixes only states with different rotational angular momenta. We decompose X into two terms:

$$X=X_{1,v_\Sigma}+X_2=C_\Sigma\langle B^1\Sigma_u^+,v_\Sigma=14,J=1,K=0|V_w|B^1\Sigma_u^+,v_\Sigma=14,K=1,I=0\rangle$$
$$+C_\pi\langle B^1\Sigma_u^+,\ v_\Sigma=14,\ J=1,\ K=0|V_w|C^1\pi_u^+,\ v_\pi=3,\ K=1,\ I=0\rangle_0, \tag{7}$$

where the subscript 0 denotes the unperturbed wave functions. As is shown above, the matrix element $X_{1,v}$ is determined by the second term in the potential V_w [Eq. (5)], which depends on the momentum of the nucleus, and it can therefore be ignored compared with the second term. In accordance with the Franck–Condon principle, we represent the matrix element X_2 in the form of a product of the electron part and the overlap integral of the vibrational wave functions:

$$X_2=C_\pi X_{2e}q_{14,3}=C_\pi\langle B^1\Sigma_u^+,\ J=1,\ K=0,\ I=1|V_w|C^1\pi_u^+,\ K=1,\ I=0\rangle_{\bar R}\langle v_\Sigma=14|v_\pi=3\rangle. \tag{8}$$

As $\bar R$, we must take the value of R whose neighborhood makes the maximal contribution to the overlap integral $q_{14,3}$. Since the left-hand "classical" turning points for these vibrational levels virtually coincide[12]:

$$r_{min}(v_\pi=3)=1{,}36\ \text{au},\qquad r_{min}(v_\Sigma=14)=1{,}31\ \text{au},$$

the maximal contribution to $q_{14,3}$ is given in accordance with the Franck–Condon principle by the neighborhood of this point.

The wave function of the excited state $C^1\pi_u^1$ was constructed by means of the simplest wave functions of Heitler–London type:

$$\psi_{\pi^\pm}{}^e=N_\pi(1s(r_{1a})2p\pi_\pm(r_{2b})+1s(r_{1b})2p\pi_\pm(r_{2a})+(1\leftrightarrow 2)). \tag{9}$$

The subscripts ± appended to the function $2p\pi$ indicate the sign of the projection of the orbital angular momentum of this atomic orbital. The required wave function can be expressed as a linear combination of the functions (9):

$$|C^1\pi_u^+,K=1,M,I=0\rangle=2^{-1/2}(\tilde D^K_{M,1}\psi^e_{\pi^+}+\tilde D^K_{M,-1}\psi^e_{\pi^-}).$$

The functions $\tilde D^K_{M\Lambda}$ depend on the spherical coordinates θ and φ of the vector R and can be expressed in terms of Wigner's d functions:

$$\tilde D_{M\Lambda}{}^K(\theta,\varphi)=\left(\frac{2K+1}{4\pi}\right)^{1/2}e^{-iM\varphi}d_{M\Lambda}{}^K(\cos\theta).$$

As wave function of the state $B^1\Sigma_u^+$, we use the wave function of Ref. 13. The matrix element $X_{2,e}$ was calculated for several values of R in the neighborhood of the turning point ($R=1.4$–1.6 au). The necessary arguments of the exponentials were calculated by means of linear interpolation between the points known from Ref. 13. It was found that $X_{2,e}$ depends weakly on R in this neighborhood, its value being approximately $X_{2,e}=3.2\times10^{-29}$ erg. To estimate $q_{14,3}$, we use the results of Refs. 8 and 9. To calculate the Λ doubling of the $C^1\pi_u$ levels in these papers, it was necessary to calculate the matrix elements of the operator $H_{\Sigma\pi}$ describing the interaction of the levels Σ and π. The matrix elements of this operator cn be represented in the form

$$H_{ij}=-\frac{(K(K+1))^{1/2}}{\mu_0}\left\langle v_\Sigma=i\left|\frac{S(R)}{R^2}\right|v_\pi=j\right\rangle,$$

where $S(R)$ is the matrix element of the electron part of the operator $H_{\Sigma\pi}$. In the pure precession approximation, $S(R)=1$. Then in accordance with the Franck–Condon principle, H_{ij} has a representation analogous to the representation (8):

$$H_{ij}=-\frac{(K(K+1))^{1/2}}{\mu_0}\frac{1}{\bar R^2}q_{ij}.$$

The matrix element of $H_{14,3}$ in the pure precession approximation was calculated in Ref. 9: $H_{14,3}=-13.1$ cm^{-1}. Taking $\bar R=1.45$, we obtain $q_{14,3}=0.08$ and $X_2=2.3\times10^{-30}$ erg. Assuming that the energy interval between the considered levels is $\Delta E=7.7$ cm^{-1}, we obtain for the coefficient of mixing of levels of opposite parity $\delta=X/\Delta E=1.5\times10^{-15}\varkappa$, which corresponds to degree of circular polarization

$$\mathscr{P}=2\delta\left[\frac{W(E1)}{W(M1)}\right]^{1/2}\varkappa\sim3\cdot10^{-5}\varkappa. \tag{10}$$

Thus, the probabilities of para–ortho transitions induced by left- or right-handed polarized radiation differ by the relative amount ~3 × 10^{-5} (if it is assumed that $\varkappa\sim1$). Therefore, the numbers of orthohydrogen molecules in cuvettes illuminated by left- and right-handed polarized light will differ by the same relative amount. As we have said above, becuase of the strong forbiddenness of the natural para–ortho transition, the relationship between the numbers of molecules of the two modifications of hydrogen can be used as a kind of memory device to accumulate information about the weak interaction. A feature of the experiment is the possibility of separating in time the stage of accumulation of information and the stage in which this information is "read out", i.e., the accumulated difference between the numbers of molecules in the two cuvettes is detected. This detection can be done by means of the resonance fluorescence signal corresponding to the allowed $E1$ transition, whose probability is ~10^8 sec^{-1}.

One can show that detection of the number of molecules of orthohydrogen with an error ~10^{-5} does not present great difficulties. Each molecule of orthohydrogen in the resonance light flux becomes a source of up to ~10^8 scattered photons in each second. Therefore, to achieve the required accuracy, it is sufficient to use a source of $\lambda \sim 1110$ Å laser radiation of power 10^{-8} W (the wavelength λ can be varied depending on the particular transition chosen for detection). However, to make ~10^{10} molecules go over from the para to the ortho state, a powerful source of laser radiation is required. We estimate the minimal power of the laser, taking ~5 days as a realistic length of the experiment.

If gaseous hydrogen is illuminated by light of frequency corresponding to the forbidden $M1$ transition,

$$^1\Sigma_g^+,\ v=0,\ K=0 \to B\,^1\Sigma_u^+,\ v=14,\ J=1,\ K=0,\ I=1,$$

then the majority of the photons will be absorbed in the wing of the adjacent intense line of the allowed $E1$ transsition. Indeed, by virtue of the Breit–Wigner formula for the absorption line profile the probability of absorption far from the resonance is determined by

$$W_{E1}'=W(E1)\,\eta,$$

where $W(E1)$ is the absorption probability in the resonance, $\eta=(\Gamma/\Delta\nu)^2$, Γ is the natural width of the absorption line, and $\Delta\nu$ is the spectral distance from the resonance frequency (the Doppler broadening can be ignored, since in this case $\Gamma_D/\Delta\nu<0.1$, $\Gamma \sim 10^9$ sec^{-1}, $\Delta\nu = 7.7$ cm^{-1}).

Since $W(E1)/W(M1) \sim 10^{20}$, the intensity of the nonresonance background absorption appreciably exceeds the intensity of the investigated para–ortho transition. Experimental conditions under which virtually all photons from the laser beam are absorbed in the sample are optimal. These conditions can be ensured by the choice of the size of the sample and the gas pressure. Under optimal experimental conditions, the number of molecules induced to go over to the ortho state can be estimated from

$$N^{n\to o} \approx \Phi t W(M1)/W(E1)\,\eta, \tag{11}$$

where Φ is the flux of photons in unit time, and t is the time of one cycle of the experiment. On the other hand, to observe parity nonconservation effects, we need to induce to go over to the ortho state not less than

$$N^{n\to o} \approx \mathscr{P}^{-2}=10^9 \text{ molecules.} \tag{12}$$

Comparing (10), (11), and (12), we obtain the required estimate:

$$\Phi \approx \frac{\eta}{4\delta^2}\frac{1}{t}. \tag{13}$$

Thus, as we have said above, our estimate of the viability of the experiment does not depend on the probability of the $M1$ transition. For the considered transition, the background level is identical with the admixed level, and therefore (13) simplifies even further:

$$\Phi \approx \frac{\Gamma^2}{4\langle V_w\rangle^2}\frac{1}{t}.$$

Therefore, the estimate of Φ does not depend on the energy gap between the ground-state level and the admixed level. Taking $t \sim 5$ days, we obtain $\Phi \sim 5\times 10^{18}$ photons/sec, which at the given frequency is ~10 W.

In recent years, laser technology has been making rapid advances in the ultraviolet region, Several methods have been developed for obtaining ultraviolet laser radiation: systems for frequency multiplication of high-power lasers by means of nonlinear media, summation of the frequencies of dye lasers, and the use of excimer molecules, consisting of atoms of inert gases and halogens. The last method is the most promising, since the excimer lasers have a high efficiency and high radiation power.[14] For example, Ref. 15 describes an ultraviolet laser using KrF molecules with pulse repetition frequency up to 1 kHz and mean power 10W. The wavelengths generated by the excimer lasers depend on the inert gas and the halogen that are used. The luminescence bands of the excimer molecules are uniformly broadened, which makes it possible to obtain a smooth tuning of the radiation frequencies.

The rotation–vibration structure of the terms of the hydrogen molecule is fairly rich and we have at our disposal numerous possible parity nonconservation effects. In addition, great interest attaches to the investigation of D_2 and T_2 molecules. A reliable calculation of the manifestation of the weak effects of the electron–nucleon interaction is possible on account of the simplicity of the molecules and the large body of both experimental and theoretical results that have been accumulated on hydrogen and its isotopic analogs.

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