PHYSICAL REVIEW LETTERS

VOLUME 21 | 30 SEPTEMBER 1968 | NUMBER 14

OBSERVATION OF LAMB SHIFTS BY QUANTUM BEATS*

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(Received 22 August 1968)

The technique which we used previously to observe quantum beats due to coherent electron-impact excitation of Zeeman magnetic sublevels is extended to the observation of the Lamb shift in the $n=4$ state of $^4He^+$. The quantum beats in our new experiment are due to temporal variations of the fluorescent light. We extend Series' theory and report the first experimental observation of the effect predicted by Series.

In 1964, Series[1] proposed measuring the energy interval between two atomic energy levels of different parity (Lamb shift) by investigation of modulated fluorescent light. He applied his idea to the $2S$-$2P$ energy interval in the hydrogen atom, in which the $2S$ state is a metastable state. This Letter reports the confirmation of Series' proposal, along with a sketch of an extension of Series' theory for the more general case with which both S and P states decay very rapidly but with different rates.

In the past, we felt that experimental confirmation of Series' proposal was very difficult, and made no attempt. However, a few months ago, we came across some very interesting experiments performed by Bashkin[2] and his collaborators, in which they observed modulation of some of the resonance fluorescence lines in He^+ and H. The excited states of He^+ and H are created by passing a fast-moving He^+ or H^+ ion, accelerated by a Van de Graaff generator, through a thin foil. A theory was constructed by Wangsness,[3] quite independent of Series, to account for Bashkin's experiment. Although the theory did not agree well with Bashkin's experimental result, we felt that the basic idea of Wangsness (hence that of Series) was quite correct. Bashkin's very interesting observation of the light-beam phenomena led to our reconsideration of Series' proposal, and, by simple extension of Series' calculation, we came to the conclusion that, by use of an electron impact method,[4] the nS-nP energy interval of He^+ and He^+-like ions must be observable by means of a quantum-beat technique.

In Bashkin's experiment, in order for the light beats to occur in the resonance fluorescence, it is necessary that, at the instant of excitation, as the ion passes through a thin foil, one of the states must be preferentially excited in comparison with another state of opposite parity. At present, there is no theory that accounts for this type of phenomenon.

On the other hand, for electron-impact ionization by the simultaneous double-excitation process, theory implies that the excitation cross section $\sigma(4S)$ for the $n=4$ S state of He^+ is smaller than $\sigma(4P)$.[5] Even if the theory is wrong, it is very likely that $\sigma(4S) \neq \sigma(4P)$. Hence the observation of the $4S$-$4P$ interval by the light-beat technique, using electron-impact ionization, offers an independent confirmation of Wangsness' explanation of Bashkin's experiment. The $n=4$ state of He^+ was chosen, since this state has been studied in detail by Lea, Leventhal, and Lamb at Yale, and Beyer and Kleinpoppen at Tübingen, using a conventional rf resonance technique.[6] The aim of our experiment was to explore the new methods of measuring Lamb shifts, and to explain the mechanisms behind Bashkin's experiments by using a completely different type of experimental method.

The experiment was performed in the following way: The electron beam was modulated at a frequency f chosen so that

$$\left(\frac{|V_{ab}|^2|\gamma_b-\gamma_a|}{\frac{1}{4}(\gamma_b-\gamma_a)^2+W_{ab}{}^2}\right) \ll f \ll \tfrac{1}{2}|\gamma_b-\gamma_a|,$$

where γ_a and γ_b are the decay constants of unperturbed $4S$ and $4P$ states, respectively; W_{ab} is the energy interval between the states represented by $|4^2S_{1/2}m\rangle$ and $|4^2P_{1/2}m'\rangle$, which can be varied by changing the externally applied magnetic field; and V_{ab} is the Stark shift, given by $V_{ab} = \langle a|\mathcal{H}_E|b\rangle/\hbar$, where $\mathcal{H}_E = -\vec{p}\cdot\vec{E}$. When the abovementioned condition for the electron-beam modulation frequency is satisfied, the amplitude of the light modulated at the frequency f goes through a significant change as $W_{ab}\to 0$ (level crossing). The existence of this phenomenon is sufficient to show the correctness of Wangsness' explanation of Bashkin's experiments.[2]

The principle behind the experiment can be explained in the following simple way: Let the states $|a\rangle$ and $|b\rangle$ be the pair of eigenstates of the atomic Hamiltonian describing the $n=4$ state of He^+ for $|4^2S_{1/2}m\rangle$ and $|4^2P_{1/2}m'\rangle$, respectively, with damping constant γ_a for the $4S$ state and γ_b for the $4P$ state. We assume that in the presence of a magnetic field the energy interval between the $|4^2S_{1/2}m\rangle$ and $|4^2P_{1/2}m'\rangle$ state is smaller than for any other pair having opposite parity. Then, in the presence of the static electric field, the time-dependent wave function $|\psi(t)\rangle$ may be expressed as a superposition of these two states, given by

$$|\psi(t)\rangle = A_0(t)\exp(-ik_a t)|a\rangle + A_1(t)\exp(-ik_b t)|b\rangle, \tag{1}$$

where $k_a\hbar = E_a$ and $k_b\hbar = E_b$, the energy of states $|a\rangle$ and $|b\rangle$, respectively. Introducing the diagonal damping Hamiltonian $\mathcal{H}_D$ defined by $\langle a|\mathcal{H}_D|a\rangle = -\frac{1}{2}i\hbar\gamma_a$ and $\langle b|\mathcal{H}_D|b\rangle = -\frac{1}{2}i\hbar\gamma_b$, and writing the total Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_E + \mathcal{H}_D$, where $\mathcal{H}_0$ is the atomic Hamiltonian in the presence of a static magnetic field, applied to reduce the energy interval between the states $|a\rangle$ and $|b\rangle$ so that the simple two-level theory can be used, and $\mathcal{H}_E$ is the Stark-shift Hamiltonian, we can readily solve the time-dependent Schrödinger equation. By solving the time-dependent equation with the initial condition that $A_0(t_0)=0$ and $A_1(t_0)=1$ for simplicity of discussion, since $\sigma(4P) > \sigma(4S)$ as implied by Dalgarno,[5] we consider the spontaneous electric dipole transition to the $n=3$ state associated with visible photons at 4686 Å. Letting $|g\rangle$ represent the $n=3$ states in which the spontaneous emission from $|\psi(t)\rangle$ is allowed, i.e.,

$$|g\rangle = \sum_{m,m',m'',m''',m''''}\{C_m|3^2S_{\frac{1}{2}}m\rangle + C_{m'}|3^2P_{\frac{1}{2}}m'\rangle + C_{m''}|3^2P_{\frac{3}{2}}m''\rangle + C_{m'''}|3^2D_{\frac{3}{2}}m'''\rangle + C_{m''''}|3^2D_{\frac{5}{2}}m''''\rangle\},$$

where the C's are the vector-coupling coefficients, one can obtain an expression for the intensity of the light radiated, $I(t,t_0)$, at time t, when the initial state $|\psi(t_0)\rangle$ was formed at $t=t_0$. This is

$$\begin{aligned} I(t,t_0) &= \frac{c}{16\pi}[\vec{E}^*\times\vec{H}+\vec{E}\times\vec{H}^*] \propto |\langle g|\hat{e}_\lambda\cdot\vec{P}|\psi(t)\rangle|^2 \\ &= K_1\{[|V_{ab}|^2/(p^2+q^2)]\{\exp[-(\tfrac{1}{2}\Gamma+p)(t-t_0)] + \exp[-(\tfrac{1}{2}\Gamma-p)(t-t_0)] - 2\cos q(t-t_0)\exp[-\tfrac{1}{2}\Gamma(t\ \ t_0)]\}\} \\ &\quad + K_2\{[4(p^2+q^2)]^{-1}\{[(\tfrac{1}{2}\delta-p)^2+(W_{ab}-q)^2]\exp[-(\tfrac{1}{2}\Gamma-p)(t-t_0)] \\ &\quad + [(\tfrac{1}{2}\delta-p)^2+(W_{ab}+q)^2]\exp[-(\tfrac{1}{2}\Gamma-p)(t-t_0)]\} \\ &\quad + 2(\alpha\cos qt-\beta\sin qt)\exp[-\tfrac{1}{2}\Gamma(t-t_0)]\} + \text{interference term}, \end{aligned} \tag{2}$$

where $\delta = \gamma_b - \gamma_a$ and $\Gamma = \gamma_b + \gamma_a$. K_1 and K_2 are the proportionality constants dependent on the polarization, oscillator strengths, etc., that describe the intensity amplitudes of the emitted light, and are of no concern to us here. The optical interference term of Eq. (2) oscillates, in general, very rapidly compared with q, the perturbed energy interval between $|a\rangle$ and $|b\rangle$, and hence one can show that this term integrated over the duration of the electron-beam pulse must vanish. The quantities q, p, α, and β are given by

$$p^2=\tfrac{1}{2}\left([\tfrac{1}{4}\delta^2-W_{ab}^{\;2}-4|V_{ab}|^2]+[\{\tfrac{1}{4}\delta^2+W_{ab}^{\;2}+4|V_{ab}|^2\}^2-4|V_{ab}|^2\delta^2]^{1/2}\right),$$

$$q^2=\tfrac{1}{2}\left(-[\tfrac{1}{4}\delta^2-W_{ab}^{\;2}-4|V_{ab}|^2]+[\{\tfrac{1}{4}\delta^2+W_{ab}^{\;2}+4|V_{ab}|^2\}^2-4|V_{ab}|^2\delta^2]^{1/2}\right),$$

and

$$\alpha+i\beta=[(p^2-\tfrac{1}{4}\delta^2+q^2-W_{ab}^{\;2})+i(q\delta-2pW_{ab})]/4(p^2+q^2).$$

These expressions agree with Series' results, if $\gamma_a=0$ and $\gamma=\gamma_b$. The physical meaning of q is the energy interval between the states $|a\rangle$ and $|b\rangle$ as perturbed by the Stark effect.

Equation (2) should explain Bashkin's findings, if the simple two-level theory can be applied to his experiment, and the result should be the same as that obtained by Wangsness.[3] Equation (2) predicts that if the state $|b\rangle$ is excited by a very sharp electron-beam pulse, the resonance fluorescence decays at two different rates, and the components that oscillate at the frequency q decay at a rate given by the average of the two damping constants.

Now if the electron beam is modulated as $\epsilon(1+\cos ft_0)$, then the intensity of light radiated at time t is given by

$$\begin{aligned}I(t)&=\epsilon\int_{-\infty}^{t}I(t,t_0)(1+\cos ft_0)dt_0,\\&=K_1'\{A_1+B_1\cos ft+C_1\sin ft\}+K_2'\{A_2+B_2\cos ft+C_2\sin ft\}.\end{aligned}\qquad(3)$$

A_1, B_1, and C_1 are all proportional to $|V_{ab}|^2$; so, for the sake of simplicity in the limit $|V_{ab}|\ll\Gamma$, we ignore the first term in comparison with the second term. The most interesting physics lies in the second term, where

$$A_2=\frac{\gamma_a(\frac{1}{4}\Gamma^2+W_{ab}^{\;2})+2|V_{ab}|^2(\Gamma)}{\gamma_a\gamma_b(\frac{1}{4}\Gamma^2+W_{ab}^{\;2})+2|V_{ab}|^2(\gamma_a^{\;2}+\gamma_b^{\;2})}\to\frac{1}{\gamma_b}$$

as $|V_{ab}|\ll\frac{1}{2}\Gamma$,

$$B_2=\frac{1}{4(p^2+q^2)}\left\{\frac{[(\frac{1}{2}\delta-p)^2+(W_{ab}-q)^2](\frac{1}{2}\Gamma-p)}{(\frac{1}{2}\Gamma-p)^2+f^2}+\frac{[(\frac{1}{2}\delta+p)^2+(W_{ab}+q)^2](\frac{1}{2}\Gamma+p)}{(\frac{1}{2}\Gamma+p)^2+f^2}\right\}+\left\{\frac{\frac{1}{2}[\alpha\Gamma-2\beta(q+f)]}{\frac{1}{4}\Gamma^2+(q+f)^2}+\frac{\frac{1}{2}[\alpha\Gamma-2\beta(q-f)]}{\frac{1}{4}\Gamma^2+(q-f)^2}\right\},$$

and

$$C_2=\frac{1}{4(p^2+q^2)}\left\{\frac{[(\frac{1}{2}\delta-p)^2+(W_{ab}-q)^2]f}{(\frac{1}{2}\Gamma-p)^2+f^2}+\frac{[(\frac{1}{2}\delta+p)^2+(W_{ab}+q)^2]f}{(\frac{1}{2}\Gamma+p)^2+f^2}\right\}+\frac{\frac{1}{2}[2\alpha(q+f)+\beta\Gamma]}{\frac{1}{4}\Gamma^2+(q+f)^2}+\frac{\frac{1}{2}[2\alpha(q-f)+\beta\Gamma]}{\frac{1}{4}\Gamma^2+(q-f)^2}.$$

Equation (3) predicts the intensity $I(t)$ of the resonance fluorescence, when the incident electron beam is modulated as $1+\cos ft$, for any arbitrary frequency f. For example, if $q=\pm f$, and $f\gg\Gamma$, the coefficients B_2 and C_2 show a resonance behavior quite similar to that of the light-beat phenomena we reported previously.[7] On the other hand, if we consider a case in which

$$|V_{ab}|^2\delta/(\tfrac{1}{4}\delta^2+W_{ab}^{\;2})\ll f\ll\tfrac{1}{2}\delta,\qquad(4)$$

the amplitude of oscillation B_2 plays the major role, and $C_2\ll B_2$ while A_2 remains constant. Under such conditions,

$$I(t) \propto \left\{ \frac{1}{\gamma_b}\left(1-\frac{|V_{ab}|^2}{\frac{1}{4}\delta^2+W_{ab}^{\;2}}\right) - 3\gamma_a \frac{\frac{1}{4}\gamma_b^{\;2}-W_{ab}}{(\frac{1}{4}\gamma_b^{\;2}+W_{ab}^{\;2})^2}\,\frac{1}{(\frac{1}{4}\delta^2+W_{ab}^{\;2})}\right\}\cos ft$$

$$\approx \frac{1}{\gamma_b}\left(1-\frac{|V_{ab}|^2}{\frac{1}{4}\delta^2+W_{ab}^{\;2}}\right)\cos ft \quad \text{when } \gamma_b > \gamma_a, \tag{5}$$

showing the variation in $I(t)$ as $W_{ab} \to 0$. This is the theoretical prediction we wished to test experimentally, showing that the S-P crossing point can be detected very accurately by observing the change in the amplitude of the $\cos ft$ oscillator term.

Figure 1 shows the experimental arrangement, which is quite self-explanatory. An electron gun having a frequency response well above a GHz was constructed to study the general form of our prediction [Eq. (3)], in addition to studying Eq. (5) resulting from the condition (4). This latter situation requires only a very low-frequency response of a few MHz. The well-evacuated electron gun, capable of maintaining a sealed-off vacuum pressure of 2×10^{-9} Torr for an indefinite period of time by the pumping action of a titanium sublimation pump, was filled with about 0.5 μ of He and placed inside an electromagnet where the magnetic field can be changed by means of a simple, operational amplifier system. A 100% modulated electron beam with a low rms current of about 1 mA/cm^2 and 200-eV energy was used in order to minimize the electric field created by the electron beam and the He^+ ions. Figure 2 shows a typical one-sweep signal for the $|4^2S_{1/2}m=-\frac{1}{2}\rangle$, $|4^2P_{1/2}m=\frac{1}{2}\rangle$ level crossing with an electron-beam modulation frequency of 2.8 MHz (for energy-level diagram, refer to Ref. 6). Since this level-crossing effect is sensitive to the electric field perpendicular to the magnetic field H, the required small electric field was caused mainly by the thermal motion of $^4He^+$ moving perpendicular to H, plus a small radial field created by the electron beam and He^+ ground-state ions. This field amounts to a few V/cm, so that the requirement of

$$|V_{ab}|^2\delta/(\tfrac{1}{4}\delta^2+W_{ab}^{\;2}) \ll f \ll \tfrac{1}{2}\delta$$

can be satisfied for $^4He^+$. A similar, but more broadened, crossing signal, with an excellent signal-to-noise ratio, was also observed for the

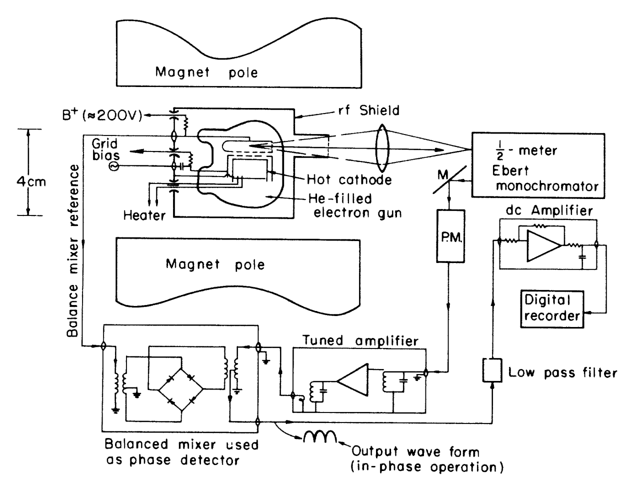

FIG. 1. Experimental arrangement used for level crossing when the electron-beam modulation frequency f is such that $|V_{ab}|^2\delta/(\frac{1}{4}\delta^2+W_{ab}^{\;2}) \ll f \ll \frac{1}{2}\delta$.

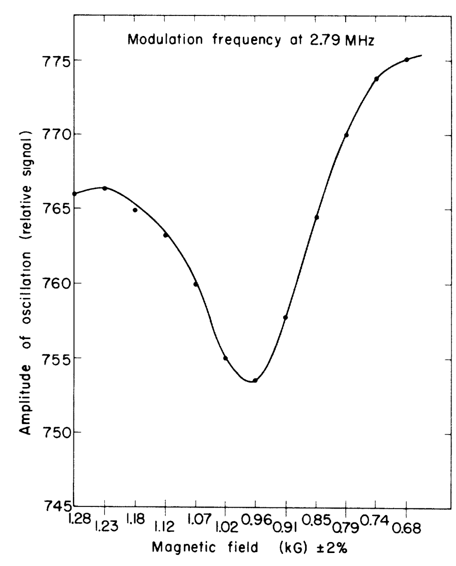

FIG. 2. Typical experimental result of the modulated intensity of resonance fluorescence as a function of externally applied magnetic field for $|4^2S_{1/2}m=-\frac{1}{2}\rangle$-$|4^2P_{1/2}\, m'=+\frac{1}{2}\rangle$ crossing point. All the experimental results showed the slope in the base line. An error of ±2 % in the magnetic field is because of the use of a flip coil that was calibrated against NMR.

$|4^2S_{1/2}m=-\frac{1}{2}\rangle$, $|4^2P_{1/2}m'=-\frac{1}{2}\rangle$ level crossing at the higher-field crossing point. In this case, the broadening was due to the electric field penetration into the screen-anode region from the control grid, as the electron gun was intentionally constructed with somewhat coarse screens to test our prediction of Eq. (3) when $f \gg \Gamma$ (order of 2-3 GHz), which requires a higher value of the electric field. This phenomenon will be reported elsewhere. Since our experimental observation is based upon Eq. (2), which also explains Bashkin's results, we believe that our experimental observations give additional support for Professor Wangsness' explanation of Professor Bashkin's experiment. Also we believe that this new method is one of the simplest ways to measure the Lamb shift in short-lived excited states.

I express my appreciation to Professor Series for teaching me about the light-beat theory, through private communications and conversations in the past. I am also grateful to Professor Bashkin for his hospitality during my visit to the University of Arizona, for showing me his experimental arrangements, and for discussions which inspired this experiment. The electron gun was constructed by Wilton Berlund of Lawrence Radiation Laboratory glass shop, and his patience in so closely following my electron gun design is greatly appreciated.

*Work supported by the U. S. Atomic Energy Commission.

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RECOILLESS RESONANCE ABSORPTION AND HYPERFINE STRUCTURE OF THE 6.2-keV STATE IN ^{181}Ta

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A strong recoilless resonance absorption was observed for the 6.2-keV level ($T_{1/2}$ $=6.8\ \mu$sec) of ^{181}Ta. Using sources of ^{181}W in W-metal and Ta-metal absorbers, a linewidth of 11 times the natural width was obtained, with isomer shifts in the range between 0.83 and 0.94 mm/sec, depending on the quality of the sources. From the hyperfine splitting in a longitudinal magnetic field the magnetic moment of the 6.2-keV level was determined to be $\mu_1=+(5.14\pm0.15)\mu_N$.

Ever since the discovery of the recoilless resonance, a considerable effort was devoted to improve the resolution of the method. The 6.2-keV transition in ^{181}Ta is one favorite candidate for this purpose, since the 6.8-μsec half-life of this level implies an extremely narrow relative linewidth,[1] $\Gamma_0/E=1.1\times10^{-14}$, and the recoil-free fraction is approximately 1 at room temperature. On the other hand, the narrow linewidth and the high spin of the transition renders the resonance extremely vulnerable even to minor interactions. Only strongly broadened resonance lines have indeed been found by earlier investigators.[2-4] The difficulties encountered are presumably due to the high sensitivity of the 6.2-keV transition against quadrupole interactions: The ^{181}Ta nucleus exhibits sizable quadrupole moments in both nuclear states, and it is difficult to produce Ta metal with a uniform cubic symmetry throughout the crystal.

We report here on a major improvement in the measurement of the 6.2-keV resonance in Ta^{181}. With the best source-absorber combinations absorption lines of about 11 times the natural width have been achieved; see Fig. 1(a). Such a narrow width allowed us to determine the magnetic mo-