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DETECTION OF SINGLY STIMULATED TWO-PHOTON EMISSION FROM METASTABLE DEUTERIUM ATOMS[*]

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The observation of singly stimulated two-photon emission from metastable deuterium atoms is reported. A collimated beam of metastables is produced by charge exchange between cesium vapor and 5-keV deuterons. A pulsed Nd-glass laser is used for the stimulation. The generated 1373-Å photons are detected at the exit slit of a spectrometer. Single-photon counting techniques are employed. The results obtained are in good agreement with theoretical calculations.

The purpose of this Letter is to report the observation of singly stimulated two-photon emission from a metastable atomic state.

The most probable mode of decay of the unperturbed metastable 2S state of hydrogen is the spontaneous two-photon decay to the ground state with a decay time of 1/7 sec.[1] The experimentally established lower limit[2] of the decay time is 1.6×10^{-3} sec. All other modes of spontaneous decay are orders of magnitude slower. The direct observation of spontaneous two-photon emission from a 2S metastable state was first reported some five years ago for He II[3] and later for Ne IX.[4]

In the presence of intense electromagnetic radiation of an optical frequency ω_{k_0}, a stimulated process (which will be referred to as singly stimulated two-photon emission) becomes significant.[5] In this process, the metastable atom decays by emitting a photon identical to and in phase with the incident photon, and a second photon of frequency $\omega_k=\omega_{2S}-\omega_{1S}-\omega_{k_0}$. The second photon is emitted spontaneously, and if the incident radiation is unpolarized, its angular distribution is given by $1+\cos^2\theta$, where θ is the angle between the photon wave vectors $\vec{k}_0$ and $\vec{k}$. For hydrogen metastables ($\hbar\omega_{2S}-\hbar\omega_{1S}=10.19$ eV) and a Nd-glass laser ($\hbar\omega_{k_0}=1.17$ eV), the wavelength of the second photon is $\lambda_k=1373$ Å (or $\hbar\omega_k=9.03$ eV) which is in the vacuum-ultraviolet range of the spectrum.

The differential cross section for the emission of the second photon $\vec{k}$ within the solid angle $d\Omega_{\vec{k}}$ is given by[5]

$$\frac{d\sigma}{d\Omega_{\vec{k}}}=r_0^{\,2}\frac{\omega_k}{\omega_{k_0}}\left(\frac{m}{\hbar}\right)^2\left|\sum_b\omega_k\omega_{k_0}\left[\frac{(\vec{r}_{1S,b}\cdot\vec{\epsilon}_{\vec{k}_0\lambda})(\vec{r}_{b,2S}\cdot\vec{\epsilon}_{\vec{k}\lambda})}{\omega_k+\omega_b-\omega_{2S}}+\frac{(\vec{r}_{1S,b}\cdot\vec{\epsilon}_{\vec{k}\lambda})(\vec{r}_{b,2S}\cdot\vec{\epsilon}_{\vec{k}_0\lambda})}{\omega_{k_0}+\omega_b-\omega_{2S}}\right]\right|^2, \tag{1}$$

where r_0 is the classical electron radius, m the electron mass, and $\vec{\epsilon}_{\vec{k}\lambda}$ the photon polarization vector. The symbol $\vec{r}_{1S,b}$ is an abbreviation for the matrix element $\langle 1S|\vec{r}|b\rangle$ and the sum is over all states of the atom. The presence of real intermediate states between the metastable and the ground state is not necessary for the process to occur. Their presence, however, may increase the cross section if such levels are in near resonance with the frequency of the incident photon. If the resonance is exact (or, strictly speaking, within the linewidth), the metastable will decay via a cascade transition which is a succession of two single-photon decays[6] and not a two-photon process in the sense used in this Letter. In the present case, the only level between $2S$ and $1S$ is the Lamb-shifted $2P_{1/2}$ level whose energy is far from resonance. Actually, the main contribution to the cross section comes from the $3P$ state which lies above the $2S$ and acts as a virtual intermediate state.

The absence of real intermediate states presents the advantage that there is no competition with single-photon transitions and, as a result, one has an unambiguous two-photon process. Also, since atomic states can shift substantially[7] under the influence of strong laser fields, real intermediate states may complicate the interpretation of similar experiments in cases of near resonance. These were among the several reasons in favor of the use of deuterium in this experiment. In addition, recent advances in the production of D($2S$) beams[8] and detection requirements in the vacuum uv made this choice almost imperative. The simplicity of the hydrogen atom and the availability of detailed calculations,[5] on the other hand, render the quantitative interpretation of the experimental results much more reliable.

The term "singly stimulated two-photon emission" was introduced in order to distinguish it from the "doubly stimulated two-photon emission." In the latter process, two incident photons with frequencies ω_1 and ω_2 adding up to $\omega_{2S}-\omega_{1S}$ induce the emission of two photons, each of which is identical to, and in phase with, one of the incident photons. This process, which is proportional to the correlation function of the incident radiation, offers an experimental tool for the study of the effects of the coherence properties of radiation on two-photon processes.[9] Similar effects have already been studied theoretically[10,11] as well as experimentally.[12]

The absence of real intermediate states and the quantitative comparison with theoretical calculations constitute two of the most important differences between the present experiment and a similar one reported by Yatsiv, Rokni, and Barak.[13] Also, both photons in Ref. 13 have optical frequencies while here the second photon is in the vacuum uv. It is felt that this is a significant difference in view of possible future applications of these processes to the creation of coherent vacuum-uv radiation.[14-16]

A pulsed Nd-glass laser was used as an intense light source. Assuming that the photon beam is essentially parallel, and denoting by I_0 its intensity measured in number of photons per second, the number of uv photons emitted per laser shot within a small solid angle $\Delta\Omega_{\vec{k}}$ is

$$R=I_0\frac{d\sigma}{d\Omega_{\vec{k}}}\Delta\Omega_{\vec{k}}Nlt, \tag{2}$$

where N is the number of metastables per cm^3 in the interaction area, l (cm) the length of the interaction area along the laser beam, and t (sec) the duration of the laser pulse.

Experimental procedures and results.—The question of experimental observation of stimulated two-photon emission from hydrogenlike atoms was first discussed by Lipeles, Gampel, and Novick[17] and later by Abella, Lipeles, and Tolk.[18] They concluded that the experiment might be more easily performed with He II metastables rather than with hydrogen because of the larger cross section and the possibility of focusing the metastable-ion beam. However, after an unsuccessful attempt to observe the effect, it became clear to us that despite these advantages, the He II experiment is at best marginal.

The observation of singly stimulated two-photon emission from deuterium atoms became feasible with the discovery that the charge-exchange reaction $d+\mathrm{Cs}\rightarrow \mathrm{D}(2S)+\mathrm{Cs}^+$ has a large cross section for deuteron energies of several keV.[8,19] This enabled us to construct a D($2S$) source which delivers a collimated beam with high current densities.

A diagram of the apparatus used is shown in Fig. 1. Deuterons are produced by an Ortec 320 rf ion source. After collimation, the 5-keV beam passes through a heated copper cell containing liquid cesium in equilibrium with its vapor at 110°C. Deuterium was chosen rather than hydrogen because of the higher cross section of the Cs charge-exchange reaction at that energy.[19]

The flux of the D($2S$) beam was measured with a calibrated 1216-Å monitor (Bendix Channeltron—tube BX 762) utilizing Stark quenching in a uniform dc electric field. In order to limit the count rate, a collimator was mounted in front of this detector. Thus only photons from a small, accurately determined part of the beam were registered. Typically 10% of the deuterons were converted to metastables resulting in a D($2S$) beam with a density of 2×10^6 metastable atoms per cm^3 and a current equivalent to 20 μA.

The beam of a 55-J pulsed Nd-glass laser was focused onto the atomic beam. With the aid of two reflectors (Al + MgF$_2$ coating, reflectivity

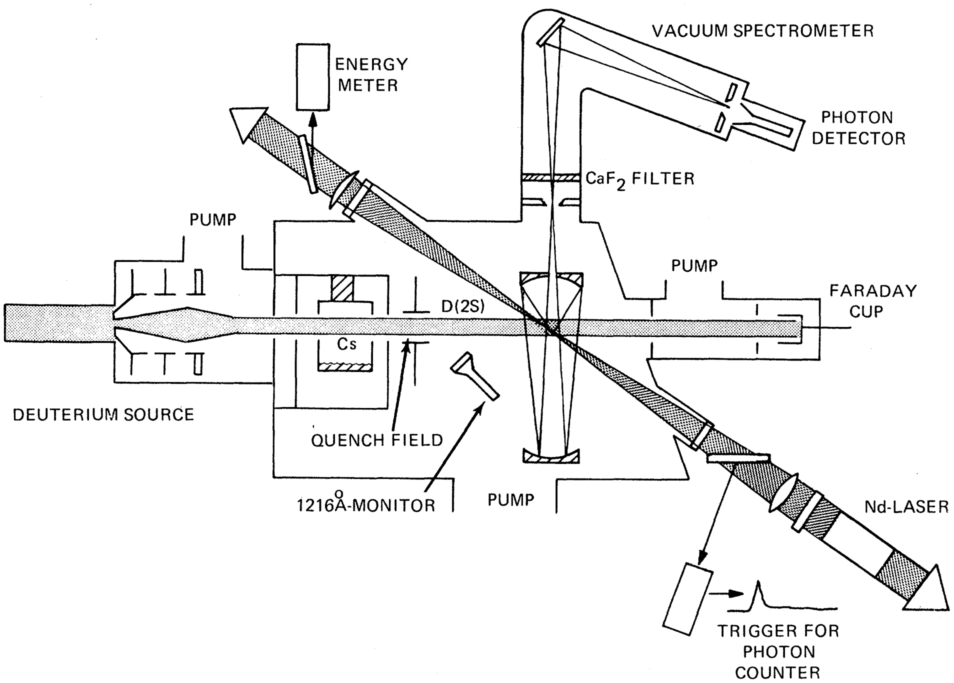

FIG. 1. Schematic diagram of experimental arrangement.

peaked at 1360 Å), the interaction area could be imaged onto the 2-mm wide entrance slit of a 50-cm Seya-Namioka vacuum uv spectrometer (dispersion 34 Å per mm). A Channeltron with a CsI-coated funnel served as the photon detector. Single photons were counted with a quantum efficiency of 25%. Considering the efficiency of the grating and accounting for its severe astigmatism,[20] fifteen 1373-Å photons were expected to be registered for 100 laser shots. This number, calculated from Eq. (2) with[5] $d\sigma/d\Omega = 1.3 \times 10^{-25}(1+\cos^2\theta)$ cm^2, is accurate only within a factor of 2, mainly due to the uncertainty in the grating efficiency and the density of metastables.

Photons were counted during the 500-μsec laser pulse and, within seconds, the background photons were registered during a time interval of equal length. In this way, the influence of possible changes of the equipment performance on the count rate was minimized. In order to assure that the difference in the count rates obtained in this fashion was indeed due to 1373-Å photons generated by singly stimulated two-photon emission, two types of control experiments were performed: (a) The metastables were quenched and the experiments described above were repeated, and (b) the laser was fired after the atom beam was interrupted.

To ascertain that the measured effect cannot be observed in other wavelength regions, all experiments were performed for different wavelength settings of the grating. Due to the required large slit width, it was necessary to cover the interval from 1290 to 1450 Å.

Per 100 laser shots or control experiments, respectively, the following number of counts were registered (grating set at 1373 Å): (a) laser beam plus background, 128.7 ± 3.7; (b) background only, 99 ± 3.2; (c) metastables quenched[21]: laser beam plus background, 100 ± 10; background only, 103 ± 10; (d) laser only, no atomic beam, 0 ± 0. The difference in the count rates from (a) and (b) decreased as expected when the spectrometer was "detuned." Below 1305 Å and above 1441 Å, no difference could be detected.

Experiments (c) and (d) assured that neither electrical pickup nor scattered photons from either the laser or the flash lamps contributed to the observed counts. A 1-mm thick CaF_2 filter prevented the detection of scattered Lyman-α photons. The results reported are based on over 2000 laser shots and an equal number of background control experiments. They were reproduced repeatedly over a period of several months.

Consequently, on the average 29.7 ± 6.9 photons of wavelength 1373 Å have been detected per 100 laser shots. In view of previously mentioned uncertainties in equipment performance, this is in good agreement with the 15 counts theoretically predicted.

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[21] Experiment (c) is based on only 100 laser shots and control experiments. Therefore, the standard deviation of these results is larger.

QUALITATIVE EXPLANATION OF PELLAM'S HELIUM PARADOX

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The temperature dependence of the torque on a Rayleigh disk immersed in rotating helium is explained qualitatively on the basis of superfluid dynamics. A heat-exchange torque peculiar to two-fluid mechanics arises due to the disk being bathed in ambient room-temperature radiation.

In the present note it will be shown that the apparently paradoxical results obtained for the torque on a Rayleigh disk immersed in rotating helium by Pellam[1] are explainable on the basis of heat torques due to ambient room-temperature radiation.

Using Landau's equations of superfluid dynamics,[2] the torque on the Rayleigh disk is given by

$$\vec{\tau} = \int \vec{r} \times (\overleftrightarrow{\pi} \cdot d\vec{S}), \tag{1}$$

where the integral is taken over the surface of the disk. $\vec{r}$ is the position vector of the surface element $d\vec{S}$ and $\overleftrightarrow{\pi}$ is the momentum-flux-density tensor given by

$$\overleftrightarrow{\pi} = \rho_n \vec{v}_n \vec{v}_n + \rho_s \vec{v}_s \vec{v}_s + p \overleftrightarrow{1}. \tag{2}$$

The torque is quite difficult to calculate in detail, but it is possible to examine the contributions to it due to two separate effects. To do so, we must realize the appropriate boundary conditions for the flow. In the first place, we assume that at the surface of the disk the normal fluid behaves as a viscous fluid, whence the parallel component of $\vec{v}_n$ vanishes.

The condition on the mass current density at