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ANTI-STOKES RAMAN SCATTERING FROM METASTABLE DEUTERIUM ATOMS*

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(Received 31 August 1970)

We have observed anti-Stokes Raman scattering from metastable deuterium atoms. The results obtained are in good agreement with theoretical calculations.

In a previous paper,[1] the observation of singly stimulated two-photon emission from the metastable $2s$ state of the deuterium atom was reported. In the presence of intense electromagnetic radiation of an optical frequency ω_{k_0}, another mode of decay of the metastable state is the emission of an anti-Stokes Raman photon. In this process, the metastable atom decays by absorbing an incident photon and emitting a photon of frequency $\omega_k = \omega_{2s} - \omega_{1s} + \omega_{k_0}$. If the incident radiation is unpolarized, the angular distribution of the emitted photons is given by $(1 + \cos^2\theta)$, where θ is the angle between the incident- and emitted-photon wave vectors $\vec{k}_0$ and $\vec{k}$, respectively. For deuterium 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 Raman photon is $\lambda_k = 1090$ Å (or $\hbar\omega_k = 11.36$ eV) which is in the vacuum ultraviolet range of the spectrum.

The differential cross section for the emission of the Raman photon $\vec{k}$ within the solid angle $d\vec{\Omega}_{\vec{k}}$ is given by[2]

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

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 states in resonance with the incident photon is not necessary for the process to occur.

If such resonant or near-resonant states do exist, however, the cross section is enhanced. In the present case, the incident photon was far from resonance with any of the intermediate states. For deuterium metastables and a Nd-glass laser, the cross section for the Raman process is about a factor of 6 larger than the cross section for singly stimulated two-photon emission.[2] However, experimental considerations subsequently discussed render its observation more difficult despite the larger cross section.

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 $\Delta\vec{\Omega}_{\vec{k}}$ is

$$R = I_0 \frac{d\sigma}{d\vec{\Omega}_{\vec{k}}} \Delta\vec{\Omega}_{\vec{k}} N l t, \tag{2}$$

where N is the number of metastables per unit volume in the interaction area, l the length of the interaction area along the laser beam, and t the duration of the laser pulse.

If, in addition to the photons ω_{k_0}, photons of frequency $\omega_k = \omega_{2s} - \omega_{1s} + \omega_{k_0}$ are present in the initial state, then the stimulated anti-Stokes Raman emission becomes possible. This process is proportional to the product of the intensities of the two frequencies, and the emitted photon is identical to, and in phase with, the incident uv photon. Provided that certain experimental conditions can be realized, it is conceivable that one may be able to use a single laser beam to generate enough anti-Stokes photons to trigger the stimulated emission. Such processes could be very valuable for the creation of coherent vacuum ultraviolet radiation.

Ducuing et al.[3] have reported the observation of anti-Stokes Raman scattering of ruby-laser light from Cr^{3+} ions in ruby. In that experiment, the emitted Raman photon had a frequency in the visible range; in the present case, the frequency is in the vacuum ultraviolet. Moreover, in our case, a quantitative comparison with theoretical calculations has been possible.

Experimental procedures and results.—The apparatus used to observe anti-Stokes Raman scattering from metastable deuterium atoms was essentially the one described in Ref. 1. A collimated beam of D(2s) atoms with a density of 2.5×10^6 metastables/cm^3 was produced from 5-keV deuterons by the charge-exchange reaction $d+\mathrm{Cs}\rightarrow \mathrm{D}(2s)+\mathrm{Cs}^+$.

A pulsed 55-J Nd-glass laser was focused onto the atomic beam, and the interaction area was imaged with the aid of a reflector system onto the 2-mm-wide slit of a 50-cm Seya-Namioka vacuum uv spectrometer. The wavelength of the Raman photons (1090 Å) is such that no suitable filter is available to suppress Lyman-α and Lyman-β photons. Since the background was expected to be extremely high, it was originally felt that the 1090-Å Raman photons could not be observed with the present experimental arrangement. However, improvements in the source of the metastables (which increased the yield to above 10%) combined with its reliable and stable performance during the work on singly stimulated two-photon emission[1] justified the attempt to observe the effect. The fact that the cross section for Raman scattering is 5.9 times larger than that for singly stimulated two-photon emission[2] was insignificant from the experimental point of view, since both the grating efficiency and the reflectivity of the Al-MgF_2 coated reflectors are much smaller at the shorter wavelength.

A minimum of 30 photons was theoretically expected to be registered with our detector system. This number was calculated from Eq. (2) and[2] $d\sigma/d\Omega=7.7\times10^{-25}(1+\cos^2\theta)$ cm^2/sr.

Single photons were counted. The quantum efficiency of the CsI-coated Channeltron electron multiplier was 20%. The accuracy of the theoretically predicted number of counted photons is only within a factor of 2, mainly because of the uncertainty in the grating efficiency and the density of metastables (see Ref. 1). This density was monitored with a specially designed Lyman-α detector[1] utilizing Stark quenching of the metastables in a dc electric field.

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, we minimized the possibility of changes in the equipment performance influencing the counting rate. In order to ensure that the difference in the counting rates obtained in this fashion was indeed due to the 1090-Å Raman photons, two types of control experiments were made: (1) The metastables were quenched to about 80% and the experiments described above were repeated. (2) The laser was fired after the atomic beam was interrupted with a mechanical shutter. Per 100 laser shots or control experiments, respectively, the following numbers of counts were registered at 1090 Å: (a) laser beam plus background, 284 ± 5.48; (b) background only, 233.5 ± 5.3; (c) metastables quenched to 80%, (1) laser plus background, 252 ± 11.2; (2) background only, 241 ± 10.7; and (d) laser only, atomic beam interrupted, 0 ± 0.

To ascertain that the measured effect cannot be observed in other wavelength regions, all experiments were repeated at different wavelength settings of the grating. Because of the large slit width, which is required for maximum intensity, the interval from 1022 to 1158 Å had to be covered.

The difference in the counting rates from (a) and (b) decreased as expected when the spectrometer was "detuned." However, at wavelengths smaller than 1060 Å, there were no conclusive experiments possible because scattered Lyman-β photons increased the background counting rate substantially. The results reported are based on over 1000 laser shots and control experiments.

We conclude that, on the average, 50.5 ± 11.1 photons of wavelength 1090 Å have been detected per 100 laser shots. In view of previously mentioned uncertainties in equipment performance, this agrees well with the 30 counts theoretically predicted.

We wish to thank Dr. R. K. Mueller for his active interest in this work and R. Hall for his assistance in performing the experiments.

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*Work supported by the U. S. Army Research Office (Durham).

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