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Observation of Radiative Collisional Fluorescence

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We report the observation of spontaneous radiative emission during the collision of two excited Ba atoms. The emission occurs at the sum energy of the excited atoms and is the emission analog of the laser-induced inelastic collision process. We measure a collision cross section for deexcitation by spontaneous radiative emission of 2.6×10^{-20} cm^2.

We report the observation of a spontaneous radiative deexcitation process, in which, during the collision of two excited atoms, a photon is spontaneously emitted at their sum energy. The process is the emission analog of a collisional absorptive process, observed some time ago in the infrared,[1] where during a collision a single photon causes the simultaneous excitation of two molecular species, causing one species to make an allowed transition, and the other a nonallowed transition.

More recently, the absorptive process has been termed a pair excitation,[2] and has been studied theoretically as a special case of the laser-induced inelastic collision processes.[3-9] Although laser-induced collisions have been observed in absorption,[10,11] we believe that this is the first report of such processes observed in spontaneous emission.

We study the process

$$\mathrm{Ba}(6p\ ^1P_1^\circ)+\mathrm{Ba}(5d\ ^1D_2) \rightarrow 2\ \mathrm{Ba}(6s^2\ ^1S_0)+\hbar\omega(3394\ \text{Å}), \quad (1)$$

where the fluorescence occurs at the sum energy of the two excited Ba atoms. Physically (and mathematically), the process may be viewed as a virtual collision, followed by a real emission. During the collision of a Ba $6p\ ^1P_1^\circ$ atom with a Ba $5d\ ^1D_2$ atom the dipole-dipole coupling causes the two atoms to make transitions to ground and to a virtual level at 29 455 cm^{-1} (with $6p'\ ^1P_1^\circ$ character), respectively (Fig. 1). While in this virtual level, the $6p'\ ^1P_1^\circ$–like atom may spontaneously emit a photon, thus resulting in the deexcitation of both atoms.

Pumping of the Ba $6p\ ^1P_1^\circ$ and $5d\ ^1D_2$ levels was obtained by two-photon excitation at $\lambda = 6614$ Å of the Ba $6p\ ^1D_2$ level (all wavelengths are given in air), followed by cascade radiative decay. The second harmonic of a Q-switched Nd-doped yttrium aluminum garnet laser was used to pump a cresyl violet dye-laser oscillator amplifier to obtain 0.8 mJ in an approximately 7-nsec-long pulse at 6614 Å. The pumping radiation was focused to an area of 3.3×10^{-3} cm^2 over 0.50 cm in the Ba heat-pipe–type cell. The cell was oper-

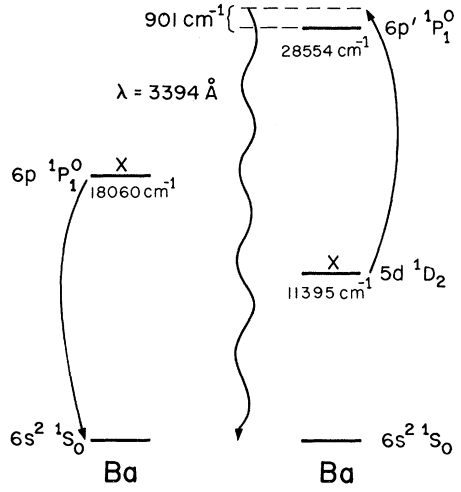

FIG. 1. Pertinent Ba levels for observation of radiative collisional fluorescence. A Ba atom in the $6p\ ^1P_1^\circ$ state collides with a Ba atom in the $5d\ ^1D_2$ state, causing the emission of a photon at the sum energy (3394 Å) and the simultaneous deexcitation of both atoms.

ated at 900°C providing a Ba vapor density of about 1.0×10^{16} atoms/cm^3, as determined by the resonance-line curve-of-growth method.[12] A noble buffer gas (either argon or xenon) was used to prevent Ba-vapor condensation on the cold cell windows.

The population in the Ba $6p\ ^1P_1^\circ$ storage level was determined by using a Chromatix flashlamp-pumped dye laser to saturate the Ba $6p\ ^1P_1^\circ$–$6p^2\ ^1S_0$ transition followed by measurement of the resulting Ba $6p^2\ ^1S_0$–$6p\ ^3P_1^\circ$ fluorescence. The Ba $5d\ ^1D_2$ population was measured by saturating the Ba $5d\ ^1D_2$–$6p'\ ^1P_1^\circ$ transition and observing Ba $6p'\ ^1P_1^\circ$–$6s^2\ ^1S_0$ fluorescence. These measurements indicated storage densities of 2.6×10^{14} atoms/cm^3 and 5.3×10^{14} atoms/cm^3 for the $6p\ ^1P_1^\circ$ and $5d\ ^1D_2$ levels, respectively. By varying the delay between the 6614-Å two-photon pump pulse and the probing pulse the effective lifetime of each of the Ba-storage levels was determined to be approximately 400± 100 nsec.

The detection system consisted of a 1-m spectrometer with a resolution of about 0.5 Å, a photomultiplier, a fast preamplifier, a pulse-height discriminator, a coincidence gate, and a counter. Typically, on line center, 230 signal counts (total counts minus background counts) were registered per 30-sec counting interval. The ratio of the signal to the standard deviation of the background was about 20.

Figure 2 shows the relative magnitude of the radiative spontaneous emission for the process of Eq. (1) as a function of wavelength. The scan covers the range between the two tabulated Ba lines at 3377.4 and 3413.8 Å.[13] These real Ba lines serve as a convenient wavelength calibration and illustrate the characteristic difference between the radiative collisional line shape at 3394 Å and the real emission lines. The plot of Fig. 2 was taken with 10 Torr of argon buffer gas. Other scans were run with 600 Torr of argon and 600 Torr of xenon. For high buffer-gas pressures the magnitudes of the real Ba lines decreased greatly, perhaps due to foreign-gas quenching; whereas the collisional emission had approximately constant magnitude and width.[10]

Spontaneous radiative emission was also observed by two-photon pumping the Ba $7d\ ^1D_2$ state

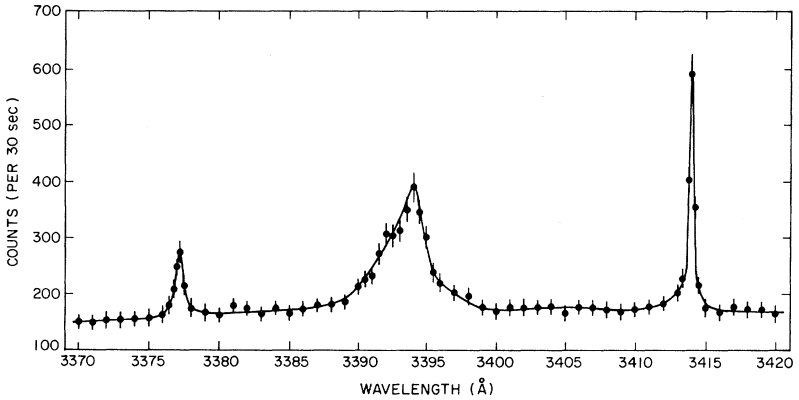

FIG. 2. Magnitude of the spontaneous radiative emission at 3394 Å with 10 Torr of argon buffer gas. Two tabulated Ba transitions at 3377.4 and 3413.8 Å serve as a convenient wavelength calibration and illustrate the characteristic difference between the line shape of a radiative collisional emission and the fluorescence from a real line. The scan has not been corrected for the 0.5-Å (~4-cm^{-1}) spectrometer resolution.

followed by radiative decay into the same storage states. The peak wavelength, and width of the emission, remained unchanged. As in Fig. 2 no unexplained emissions were observed, although the relative magnitudes of the real Ba lines were changed.

The collisional fluorescence of Fig. 2 has a maximum at the predicted ($R=\infty$) wavelength of 3394 Å and a half-power linewidth of about 20 cm^{-1}. An approximate calculation indicates that the resonant dipole-dipole interaction C_3/R^3, with $C_3=1.3\times10^{-35}$ erg cm^3, is the dominant term in the interaction energy. This interaction results in an orientationally dependent, symmetric splitting of the interaction potentials[14]; which in turn implies an approximately symmetric line shape for the radiative collisional fluorescence. If additional C_0/R^6 terms are included, an asymmetric tail is predicted.

The magnitude of the cross section for collisional deexcitation by spontaneous emission σ_s was determined by measuring the total number of photons (integrated over the emission bandwidth) at 3394 Å and using, for the number of detected photons,

$$N(\mathrm{Ba}\ 6p\ ^1P_1^\circ)N(\mathrm{Ba}\ 5d\ ^1D_2)\sigma_s\overline{V}\tau\Delta V\eta,$$

where $\overline{V}$ is the average velocity, ΔV is the effective radiating volume, η is the ratio of detected to generated photons, and τ is the effective radiating time of the media. For a typical 30-sec counting interval, the integrated signal count rate was 1465 counts. As described above, the excited Ba densities were measured by optical probing and were $N(\mathrm{Ba}\ 6p\ ^1P_1^\circ)=2.6\times10^{14}$ atoms/cm^3 and $N(\mathrm{Ba}\ 5d\ ^1D_2)=5.3\times10^{14}$ atoms/cm^3; $\overline{V}=4.3\times10^4$ cm/sec, $\Delta V\cong1.7\times10^{-2}$ cm^3, $\eta=9.4\times10^{-5}$, and $\tau=10$ pulses/sec$\times30$ sec$\times200$ nsec/pulse$=6.0\times10^{-5}$ sec. (Each storage state decays in ~200 nsec.) We thereby obtain a velocity averaged collision cross section of $\sigma_s\cong2.6\times10^{-20}$ cm^2, with an overall experimental uncertainty of approximately a factor of 5.

One may relate the measured cross section for deexcitation by spontaneous emission, and measured line shape, to two other quantities of interest. The first of these is the cross section for stimulated deexcitation of the Ba levels due to intense laser radiation applied near $\lambda=3394$ Å. This cross section, also termed the cross section for laser-induced or -switched collision, is a function of the applied laser frequency, and for deexcitation at line center is approximately

$$\sigma_c=\frac{1}{\hbar}\frac{1}{\omega^3}\frac{\pi^2c^2}{\delta\omega}\sigma_s\frac{P}{A}, \tag{2}$$

where $\delta\omega$ is the (approximate) full-width–half-power linewidth. Using $\sigma_s=2.6\times10^{-20}$ cm^2 and $\delta\omega=3.8\times10^{12}$ rad/sec, we infer $\sigma_c=3.4\times10^{-24}\times P(\mathrm{W})/A(cm^2)$ cm^2. This agrees well with a calculated value of $\sigma_c=1.9\times10^{-24}P(\mathrm{W})/A(cm^2)$ cm^2 and is of the same order of magnitude as previously measured cross sections for laser-induced collision. We see that an incident laser power density, for this system, must exceed about 2×10^4 W/cm^2 for the stimulated collisional deexcitation to exceed the spontaneous deexcitation. It should be noted that both σ_s and σ_c are enhanced by the presence of the relatively nearby $6p'\ ^1P_1^\circ$ level.[8]

The other quantity of interest which may be obtained from the measured values of σ_s and $\delta\omega$ is the absorption coefficient for the inverse pair-absorption process, or the gain coefficient for a system where the product of excited state densities exceeds the product of the ground-state densities. The absorption (or gain) coefficient may be written

$$\alpha=\frac{\pi^2c^2\overline{V}\sigma_s}{\omega^2\delta\omega}\left(\frac{g(A^*)g(B^*)}{g(A)g(B)}N(A)N(B)-N(A^*)N(B^*)\right), \tag{3}$$

where $N(A), N(B)$ and $N(A^*), N(B^*)$ are the appropriate ground- and excited-state storage densities, respectively, and $g(A)$, etc., are the corresponding degeneracies. For the Ba system studied here, for pair absorption, $\alpha=1.3\times10^{-36}N(\mathrm{Ba}\ 6s^2\ ^1S_0)N(\mathrm{Ba}\ 6s^2\ ^1S_0)$ cm^{-1}. For absorption of several percent per centimeter, densities of about 2×10^{17} atoms/cm^3 are necessary.[15]

The results of this experiment may have application to the construction of low-gain, high-energy storage laser media and to spectroscopic studies of the interaction potentials of colliding atoms.

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[15]In a recent experiment, using a white light source, we have observed such an absorption in the Ba-Ba system. The absorption peaked at the $R=\infty$ wavelength of 3394 Å and had a comparable line shape to that observed in emission.

Nuclear "Time-Delay" and X-Ray–Proton Coincidences near a Nuclear Scattering Resonance

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The probability for production of K-shell x rays by protons elastically scattered from ^{58}Ni is observed to change as one varies the proton energy over the $s_{1/2}$ nuclear resonance at $E_p = 3.151$ MeV. The results are interpreted theoretically in terms of interfering products of atomic ionization and energy-dependent nuclear scattering amplitudes.

We present experimental evidence for a change in the probability for production of K-shell x rays by protons elastically scattered to a definite angle θ as one varies the incident proton energy over a narrow nuclear resonance. This is the first experiment in which the nuclear "time delay" associated with a resonance has been observed to change the rate of an atomic excitation. In a time-dependent description, a time delay τ causes the amplitude for excitation of an electron "before" to be advanced in time relative to the amplitude for excitation "after" the nuclear collision by the factor $\exp(-i\omega\tau)$, where $\hbar\omega$ is the energy transfer to the electron.[1,2] In the time-independent description actually used in interpreting our experiment, we find that the exponential factor is replaced by the ratio of nuclear-reaction amplitudes $f(E-\hbar\omega)/f(E)$; time delay is manifested by the energy variation of these amplitudes. We demonstrate that this type of experiment, unlike previous scattering experiments,[3] is sensitive to the imaginary part of the dominant (monopole) atomic ionization amplitudes. The present experimental results suggest that this imaginary part may not be well understood.

Our theoretical treatment was motivated by the need for expressions appropriate to experiments which involve a "mixture" of nuclear lifetimes (as is the case for a resonance-plus-Coulomb scattering) and incident beams whose energy spread is small compared to the width of the nuclear resonance. Semiclassical time-dependent perturbation-theory treatments[1,2] of K-shell ionization are inappropriate in these circumstances. Therefore, we have reformulated the theory of K-shell ionization in terms of time-independent perturbation theory, taking explicit account of conservation of energy and angular momentum in all portions of the electron-nuclear wave function as well as correct matching of nuclear wave functions at a radius small compared to atomic but large compared to nuclear dimensions; a WKB approximation is used in obtaining the atomic amplitudes. The resulting cross section for producing a nuclear particle at angle θ (c.m.) in coincidence with a K-shell X ray takes the intu-

