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Inhibited Absorption of Blackbody Radiation

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A cutoff with wavelength is observed in the absorption of blackbody radiation by free atoms. The cutoff arises from a discontinuity in the density of modes between parallel conducting plates. Absorption at a wavelength of $\frac{2}{3}$ cm by atoms between planes $\frac{1}{3}$ cm apart is measured at a temperature of 180 K. The discontinuity in the absorption rate occurs when the absorption wavelength is varied across the cutoff of one of the parallel plate modes.

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The effect of conducting surfaces on the radiation rate of elementary atomic systems has been studied theoretically by a number of investigators,[1-4] but experimental evidence is scarce. The principal experimental work is an elegant series of studies by Drexhage on fluorescence of a thin dye film near a mirror.[5] Drexhage observed an alteration in the fluorescence lifetime, arising from the interference of the molecular radiation with its surface image. The radiation rate in such an experiment is sensitive to the optical properties of the surface, and quenching-rate data on surface-deposited molecules can yield detailed information on molecule-surface interactions.[6] We have undertaken a study of the radiative properties of atoms in the proximity of conductors with a somewhat different goal: to alter the coupling of atoms with the vacuum by manipulating the mode structure of the system. We report here the results of a study of blackbody radiative transfer of free atoms between conducting planes at a wavelength so long that the conductor is, to good approximation, ideal. The transfer rate undergoes a discontinuity with frequency which can be explained in terms of the effect of the elementary mode structure on the spontaneous decay rate. The experiment serves as a step toward the achievement of conditions in which spontaneous emission can be effectively eliminated.[4]

The experiment involves radiative transfer between two Rydberg states of sodium by absorption of thermal radiation at a wavelength of $\frac{2}{3}$ cm. Absorption occurs midway between parallel conducting plates $\frac{1}{3}$ cm apart. A small dc field is applied between the plates to vary the absorption wavelength by the Stark effect. The field is slowly increased, and as the wavelength drops below the cutoff value for one of the parallel plate modes, the absorption rate is seen to increase abruptly.

The rate for radiative absorption between two states is $A\bar{n}$, where A is the spontaneous transition rate and $\bar{n}$ is the photon occupation number. For a thermal field, $\bar{n}=[\exp(h\nu/kT)-1]^{-1}$. A is proportional to the mode density $\rho(\nu)$ for photons at the transition frequency ν.[3,4] In free space, $\rho(\nu)=4\pi\nu^2/c^3$. Between parallel conducting planes there are families of modes for the electric field parallel and perpendicular to a normal to the planes, $\rho_\parallel$ and $\rho_\perp$, respectively. The characteristic cutoff frequency is $\nu=c/2d$, where d is the plate separation. In the frequency region $0<\nu<2\nu_c$, the mode densities are

$$\rho_\parallel = 4\pi\frac{\nu_c\nu}{c^3},$$

$$\rho_\perp = \begin{cases} 4\pi\dfrac{\nu_c\nu}{c^3}, & \nu>\nu_c, \\ 0, & \nu<\nu_c. \end{cases}$$

The same results, starting from a somewhat different point of view, were observed previously by Milonni and Knight.[2] Absorption was observed on both modes simultaneously, and the step in the absorption rate due to the "turnon" of $\rho_\perp$ was observed at $\nu = \nu_c$.

The transition employed is $29d \to 30p$. Energy levels are shown in Fig. 1. (Energies and frequencies are in inverse centimeters.) At zero applied electric field the transition occurs at 1.44 cm^{-1}, while at 6 V/cm it is at 1.64 cm^{-1}. The cutoff frequency for the plate separation of 0.337 cm is $\nu_c = 1.48$ cm^{-1}. Thus, as the electric field is varied between 0.7 and 5.7 V/cm, the frequency varies between $0.97\nu_c$ and $1.11\nu_c$, reaching the cutoff frequency at 2.4 V/cm.

A population of $29d$ atoms in an atomic beam is prepared by stepwise excitation with two laser beams via the route $3s \to 3p \to 29d$. The atoms are excited by simultaneous 10-ns laser pulses at a 10-Hz repetition rate. Transitions to the $30p$ states are then induced by radiation from the walls of a carefully shielded enclosure maintained at 180 K.[7] The excited population interacts with the radiation field for approximately 20 μs. The spontaneous rate for the transition $29d \to 30p$ is $A(29d, 30p) = 3.5$ s^{-1} and the photon occupation number at 180 K is $\bar{n} \simeq kT/h\nu = 86$. Thus the total transfer rate at zero field is 300 s^{-1}. The final-state distribution is analyzed by selective field ionization.[8] The ionization signal includes atoms in all of the $30p$ sublevels, plus some background from higher-lying levels, but excludes atoms in the $29d$ state.

The selection rules for blackbody absorption in the two radiation modes are $\Delta m = 0$ for $\rho_\parallel$, $\Delta m = \pm 1$ for $\rho_\perp$. Because the Stark effect in the $29d$ and $30p$ states is large compared to the fine and hyperfine interactions, the selection rules operate on the orbital quantum number m_l. The initial $29d$-state populations are determined by the laser excitation process. Both lasers are linearly polarized along the z axis ($E_\parallel$). The first laser drives the transition $3s \to 3p$ with the selection rule $\Delta m_F = 0$. (The total angular momentum F must be considered because hyperfine as well as fine structure is important in the $3p$ state.) The second laser excites atoms between regimes where initially m_F is a good quantum number and finally m_l is a good quantum number. The result is that all m_l levels of the $29d$ state are populated, and all allowed transitions $29d \to 30p$ can be observed.

Experimental results are shown in Fig. 2. The abrupt increase in the transfer rate at $\nu = 1.00\nu_c$ is apparent. (Note that the width of the "step" is exaggerated by the expanded frequency scale. The fractional width is approximately 1%). The ratio of rates above and below cutoff is 1.7:1. The theoretically expected ratio depends on the distribution of populations in the $29d$ state, as well as on the spatial distribution of the atoms between the conducting plates. For the ideal case of uniformly populated $29d$ states located at the midplane, the ratio is 5:1. The fact that the observed ratio is significantly smaller than the ideal value may be due to some breakdown of these assumptions, but it is most likely due to the presence of a large background of ionization arising from radiative transfer to levels lying above the $30p$ state. To verify that the observed step in the transfer rate is indeed due to "turnon" of $\rho_\perp$, radiative transfer was studied in a number of manifolds adjacent to $29d$. Data for a typical example, transfer on the $28d \to 29p$ transition, is shown for comparison in Fig. 2. This transition lies entirely above cutoff; neither this nor any of the other adjacent transitions shows a discontinuity.

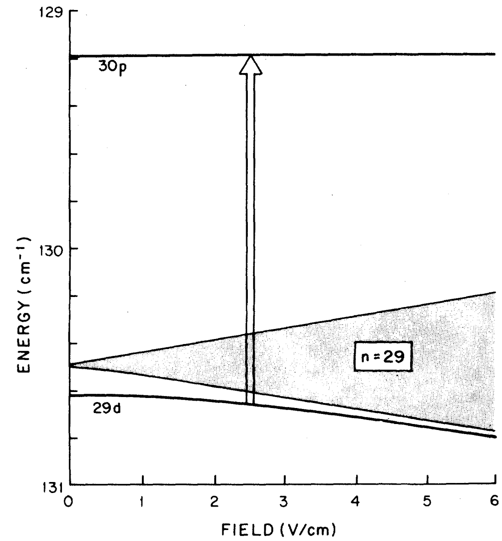

FIG. 1. Energy-level diagrams for sodium in the vicinity of $n = 29$. The diagrams for $m = 0$ and $|m| = 1$ are indistinguishable. Blackbody radiative transfer was observed on the transition $29d \to 30p$, shown by the arrow. The shaded region is the manifold of linear $n = 29$ Stark states.

We have calculated in detail the dependence of the radiative transfer rates with field for all the transitions observed. Variations in the oscillator strength distributions and in the spontaneous life-

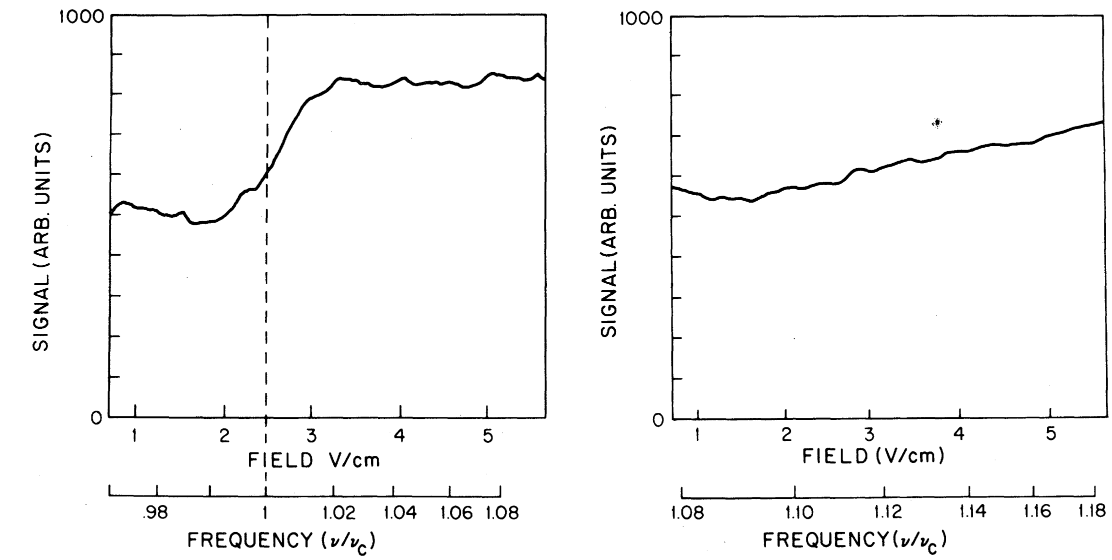

FIG. 2. Blackbody radiative transfer signals in sodium located between parallel conducting plates for $29d \rightarrow 30p$ (left-hand side) and $28d \rightarrow 29p$ (right-hand side) as a function of the absorption frequency. The critical frequency is $\nu_c = 1/2d = 1.48$ cm^{-1}, where d is the plate separation. The increase in the transfer rate at $\nu = \nu_c$ (left-hand side) is due to the "switching on" of the $\rho_\perp$ mode.

times both play important roles in determining the shapes of the transfer curves. The rates vary monotonically with the field, except for the transition $29d \rightarrow 30p$ which displays a discontinuity. Although a number of experimental difficulties have so far prevented a quantitative study of the rates over a wide range of field, we believe that the observations in the vicinity $\nu/\nu_c \simeq 1$ are reliable. In particular, the fact that the Stark field at the cutoff frequency agrees with the calculated field to within the experimental resolution, approximately 1%, provides strong evidence that the observed discontinuity is due to the cutoff in $\rho_\perp$.

We have calculated the effect on the mode distribution of the finite diameter of the conducting planes. The field plates are disks surrounded by a cylindrical copper shield 8 cm in diameter, with a radial gap of approximately 2 mm. The cylindrical wall imposes a fine structure on the parallel-plate modes with a periodicity of approximately 0.06 cm^{-1}. The fine structure is expected to be heavily damped by losses due to the radial gap, however, and we saw no evidence of it.

The connection between blackbody absorption, stimulated emission, and spontaneous emission is so intimate that observation of inhibited absorption provides compelling, though indirect, evidence for inhibited spontaneous emission. For example, the reasoning which motivated Einstein's theory of radiation in free space[9] remains valid when finite boundary conditions are imposed. Thus if the ratio of spontaneous to stimulated emission were altered from the free-space value by the conducting planes, one could devise an engine in which atoms transfer heat from a radiation field to the walls of the cavity with no other effect on the system, in defiance of the principles of thermodynamics. From this it follows that inhibited absorption necessarily implies inhibited spontaneous emission. Alternatively, one need merely point out that the same density of modes appears in the quantum electrodynamic expressions for spontaneous and stimulated radiation rates.[10] Nevertheless, the direct observation of inhibited spontaneous emission would provide new opportunities to study atom-vacuum interactions. Such experiments now appear to be feasible, and we are pursuing research in this direction. In addition, we are carrying forward radiative transfer studies in which the initial and final states can be fully resolved, permitting a quantitative study of the effect of mode structure on the transfer rate.

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Observation of the Modification of "Optical" Collision Dynamics in Intense Laser Fields

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The qualitative form of the predictions of Light and Szöke for an optical collision involving the $\lambda = 460.7$-nm transition in strontium perturbed by argon has been verified. In the presence of linearly polarized laser light the $m_J = 0$ to $m_J = 0$ is "switched off" at large field intensities ($\gtrsim 50$ MW/cm^2) whereas the $m_J = 0$ to $m_J = \pm 1$ transition is not observed to saturate. The consequences for the study of laser-induced phenomena is discussed.

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In order to determine whether molecular collision processes can be usefully modified by the presence of an intense field, theorists and experimentalists have expended considerable effort.[1,2] A relatively simple and important prototype for such studies is far-wing absorption of laser light in the presence of a collisional perturbation (an "optical collision"),[3] whose theoretical description in terms of dressed states[4] has proven most profitable. In this picture the absorption process occurs via a curve crossing[5] between the dressed upper and lower quasimolecular states. Lisitsa and Yakovlenko[6] discussed how this process would be modified in the presence of a sufficiently strong laser field and showed, in a two-state model, that the curve crossing becomes an avoided crossing; the optical collision cross section should then drop off exponentially for high laser intensities.[7]

Light and Szöke[8] pointed out a severe deficiency in the two-state model, and performed a more realistic calculation of the cross sections (using an S-matrix approach in the dressed frame) for a Zeeman-degenerate atom. They specifically treated the $\lambda = 460$-nm ($J_g = 0$ to $J_e = 1$) resonance transition of strontium collisionally perturbed by argon via a long-range van der Waals interaction and showed, for linearly polarized laser light, that the strongly coupled $m_J = 0$ to $m_J = 0$ transition would have the variation in cross section expected from the two-state case, but that the transitions to the $m_J = \pm 1$ sublevels of the excited state would not. This would have important consequences for the field of laser-induced processes as it showed that population in this system could be forced into the channels that were not strongly coupled by the incoming radiation. Fortunately, the results of Light and Szöke[8] have a clear, qualitative, experimentally observable prediction: a strong variation of the polarization of the fluorescence with laser field strength. In this Letter we present the experimental verification of the nature of the predictions of Light and Szöke.[8]

In our experiment we do not observe the variation of the optical collision cross sections directly, but rather the effect of that variation on the collisional rates, and hence on the population kinetics in the dressed frame. The formal theory of dressed-state kinetics in the presence of radiative and collisional relaxation in the steady state