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Production of "hot" excited-state atoms in collisionally aided radiative transitions

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Collisionally aided radiative excitation of atoms using a laser detuned to the blue is shown to lead to heating of the external degrees of freedom. The velocity distribution of the excited atoms is studied experimentally and compared with theoretical predictions.

The combined effect of collisional and radiative interactions upon an atom in a vapor has been the subject of a great deal of theoretical and experimental activity in the past several years.[1,2] A class of such reactions, commonly referred to as collisionally aided radiative excitation (CARE), can be described in a very general way as

$$A_1 + B_1 + h\Omega \rightarrow A_2 + B_1 \ ,$$

where A_i and B_j represent atoms of different species A and B in states i and j, respectively. Experimental studies to date have mainly concentrated on measurements of either CARE cross sections or the frequency distribution of the reemitted light. However, as implied in most recent treatments of light scattering, this type of process also produces a change in the atomic velocities. The velocity changes can be viewed very easily: Let us consider radiation of frequency Ω acting on a two-level system whose Bohr frequency is ω (with $\Omega - \omega = \Delta >>$ Doppler width). As a result of the interaction with the field, the atom may undergo Rayleigh scattering or be excited to its upper state. The latter process, although possible without collisions in the presence of a strong field, is greatly enhanced by collisions.[2,3] Since the final energy level of the atom is different from the energy of the absorbed photon, the energy difference must be compensated by a change in the translational energy of the colliding atoms.

As pointed out previously,[4,5] this process may have a potential use as a method for heating or cooling an atomic vapor. Tuning with $\Omega > \omega$ produces heating whereas that with $\Omega < \omega$ produces cooling.

We have studied the case of the sodium–rare-gas system subjected to laser irradiation detuned from the sodium $3S \rightarrow 3P$ transition toward high frequencies:

$$\mathrm{Na}(3S_{1/2}) + (\text{rare gas}) + h\Omega \rightarrow \mathrm{Na}(3P_{1/2}) + (\text{rare gas}) \ .$$

The velocity distribution of the excited $3P$ sodium atoms was monitored by scanning a narrow-band probe laser through the Doppler-broadened resonance associated with absorption on the $3P_{1/2} \rightarrow 4D_{3/2}$ transition (Fig. 1).

The rare-gas perturbers used in our experiment were helium ($M = 4$), argon ($M = 40$), and xenon ($M = 130$). The excess energy $h\Delta = h(\Omega = \omega)$ is distributed between the two partners of the collision, sodium ($M = 23$) and rare gas according to the usual laws of classical mechanics. Consequently, we expect very small velocity changes for sodium in the helium-sodium collision, since the lighter helium atom carries off practically all the excess energy $h\Delta$. The velocity distribution of the $3P$ excited sodium atom in that case will be essentially identical to the Maxwellian velocity distribution in the ground state; this provides a control measurement to which the velocity changes obtained in collision with the heavier rare gases, argon and xenon, can be compared. In the latter cases, the velocities of the $3P$ excited sodium atoms will be nonthermal and characterized by a nonequilibrium distribution $\rho(\vec{v})$ which has been calculated using standard methods[6]:

$$\rho(\vec{v}) = \frac{P}{\gamma} W_0(\vec{v}) \int W(\vec{v}' \rightarrow \vec{v}) d\vec{v}' \ ,$$

where P is the excitation probability, γ^{-1} the lifetime of the $3P_{1/2}$ level, and $W_0(\vec{v})$ the Maxwellian distribution. The collision kernel $W(\vec{v}' \rightarrow \vec{v})$ has been computed assuming a hard-sphere scattering potential. This can be justified because at the large blue detuning corresponding to our experimental situation, the main contribution comes from collisions with small impact parameters, in which the repulsive core of the interatomic potential is dominant. The longitudinal $3P$ velocity distributions are shown in Fig. 2; the $3P_{1/2} \rightarrow 4D_{3/2}$ absorption profiles have essentially the same shape, with abscissa $ku(v_z/u)/2\pi$. These curves deviate from Gaussian profiles since there are no "slow" atoms in the velocity distribution. With $\Delta = 19.09 \times 10^3$ GHz at 250°C ($h\Delta \simeq 1.6$ kT) the full widths at half maximum

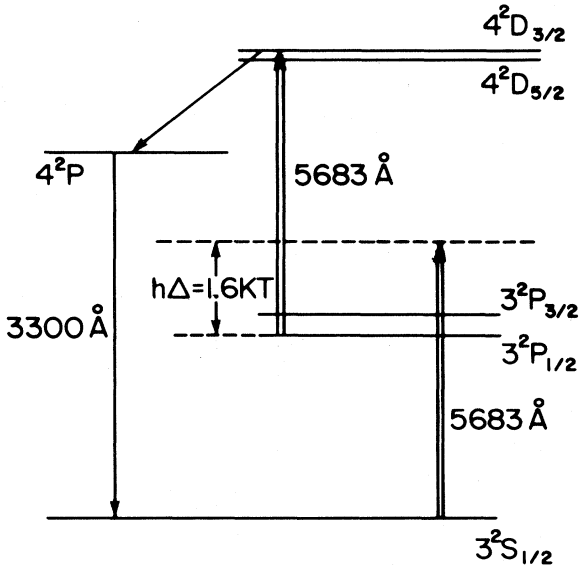

FIG. 1. Sodium energy-level diagram and excitation scheme.

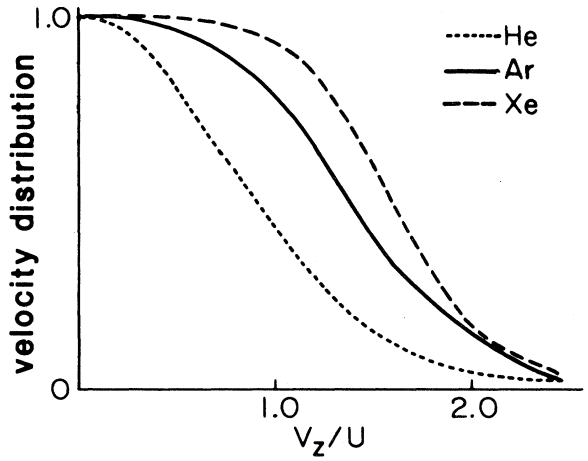

FIG. 2. Normalized theoretical velocity distributions after $3S \rightarrow 3P$ collisionally aided radiative excitation in the presence of helium, argon, and xenon. (v_z is the velocity component along the laser beam, u^2 the mean-square velocity.)

(FWHM's) are calculated to be, respectively, 1.9, 2.9, and 3.5 GHz for helium, argon, and xenon perturbers. The FWHM given by a Maxwellian velocity distribution at the same temperature is 1.8 GHz.

The experimental setup is shown in Fig. 3. The experiment was performed in an alkali-resistant glass cell connected to a vacuum system, and kept by an oven at a temperature of about 250 °C, producing a sodium density of 5×10^{13} cm^{-3}. Rare-gas (helium, argon, xenon) pressures between 5 and 30 Torr were used. The atoms were irradiated by laser light focused into the cell to a diameter of about 30 μm. The collisionally aided $3S \rightarrow 3P$ excitation and the probing on the $3P \rightarrow 4D$ transition were achieved with the same laser, tuned to the $3P_{1/2} \rightarrow 4D_{3/2}$ resonance (5683 Å). The corresponding detuning of the laser from the $3S_{1/2} \rightarrow 3P_{1/2}$ resonance (5896 Å) is 19.09×10^3 GHz. The laser used in an argon-ion-pumped single-mode ring dye laser operated with rhodamine 6G at a power of 50 mW to avoid excessive saturation of the $3P \rightarrow 4D$ transition. The laser was scanned over 30 GHz across the $3P_{1/2} \rightarrow 4D_{3/2}$ resonance. (The magnitude of this scanning range is very small compared with the frequency detuning Δ from the $3S \rightarrow 3P$ transition, and the resulting variation of Δ can be neglected.) The scan was calibrated by passing a small part

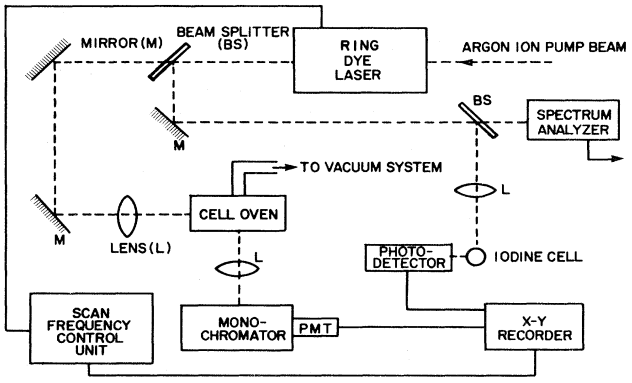

FIG. 3. Experimental setup.

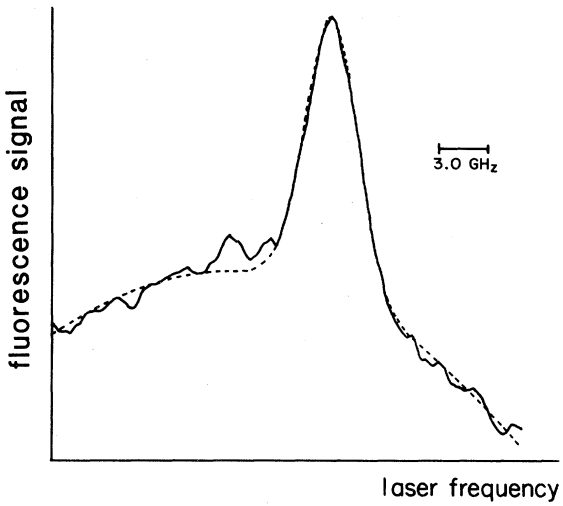

FIG. 4. Na-Xe collisional line shape recorded on the $4D \rightarrow 3P$ fluorescence (solid line). $T = 250\,°C$, $p_{Xe} = 20$ Torr. Because of the low transmission of the alkali-resistant glass in the UV, we could not use the 3300-Å line associated with the $4D \rightarrow 4P \rightarrow 3S$ cascade. The $4D \rightarrow 3P$ fluorescence appears on the background laser light, which has been taken into account by including a broad parabola in the fitting function (dashed line).

of the laser beam through an iodine cell and recording the absorption spectrum.[7] The $3P_{1/2} \rightarrow 4D_{3/2}$ transition line shape was measured by monitoring the fluorescence emitted from the $4D_{3/2}$ level on an x-y recorder or a data digitizer for computer analysis. An example of the latter is shown in Fig. 4.

As discussed above, the width of this Doppler-broadened absorption line gives a measurement of the mean speed of the $3P$ atoms along the laser propagation axis. Table I shows the FWHM measured in our experiments for various pressures of rare-gas perturbers. There is an obvious difference in the widths obtained for helium and the two heavy rare gases. However, these raw data actually include pressure and power broadening effects on the probe transition which we have extracted for a more quantitative comparison with the theoretical Doppler widths given above. This was done making use of the pressure and power broadening results obtained with helium as the perturber. The Doppler profile in that case is essentially the Gaussian curve associated with the Maxwellian distribution. The absorption line shape is a Voigt profile that results from the convolution of the Gaussian-Doppler line shape with a Lorentzian function, the width of which is determined by pressure and light-

TABLE I. Widths, in GHz, of the probe $3P \rightarrow 4D$ transition. The estimated errors are about 0.1 GHz.

Rare-gas pressure (Torr)	Na-He	Na-Ar	Na-Xe
5	2.7	3.0	
10	2.7	3.2	3.5
20	3.1	3.3	4.0
30	3.3	3.6	

power broadening (the natural width and hfs can be neglected here). Study of the dependence of the widths upon laser power shows that the power broadening is significant. However, at a laser power of 50 mW, which was used in all the experimental runs, it is rather small and is accounted for in our data processing at the same time as pressure broadening, as shown below. For the analysis of pressure broadening effects, we rely on previous experimental studies[8,9] which indicate that the pressure broadening coefficients for helium, argon, and xenon are the same within 15% for the $3P \rightarrow 4D$ transition. We have thus assumed the same Lorentzian widths for the heavy rare gases as for helium at the same pressure, and we have used the values of the Lorentzian widths experimentally determined by the deconvolution of the curves obtained with helium to interpret the absorption curves obtained with argon and xenon. We obtained the Doppler contributions to the linewidths for the latter two gases by a deconvolution also assuming a Voigt profile (this assumption will be discussed below). Thus we obtained values for the Doppler widths which are no longer dependent on the foreign gas pressure.

These are shown in Table II, along with the theoretical predictions. For both argon and xenon, it can be seen that the experimental values are significantly larger than the thermal Doppler widths. This gives evidence for the predicted "heating" effect. Nevertheless, the effect is smaller than theoretically calculated. One can account for this if one considers the processes that thermalize the atoms in the $3P$ state. First, once excited to the $3P$ state and before absorbing a second laser photon, the sodium atoms may undergo collisions with the perturbers which tend to return the velocity distribution to the thermal one. We note that the laser power density is large enough for the $3P \rightarrow 4D$ transition to be partly saturated, which reduces these velocity changes. Second, the reabsorption of resonance photons, which provides an alternative mechanism to excite atoms from the ground state, has the same effect of washing out the particular velocity distribution of the $3P$ atoms by CARE. Although we work at densities of sodium where the optical depth for resonant light is high, the transit time of an atom excited by CARE through the laser beam is small, so that reabsorption events take place mainly outside of the laser beam, where we do not detect them. These effects are very difficult to estimate quantitatively. In order to study them it would be necessary to perform experiments at lower sodium and rare-gas pressures, which we were unable to do because of signal-to-noise ratio considerations. The partial thermalization in the $3P$ level may also explain why the experimental absorption line shape (Fig. 4) for the $3P \rightarrow 4D$ transition differs from the theoretically predicted one (Fig. 2), and justifies our use of a Voigt profile in fitting the data.

TABLE II. Comparison of the experimentally obtained widths with theory (power and pressure broadening extracted). All values are in GHz.

Gas	Thermal Doppler width	Theoretical width including heating	Experimental Doppler width
Ar	1.8	2.9	2.4
Xe	1.8	3.5	2.8

In conclusion, our experiment has shown evidence for significant "heating" associated with the collisionally aided radiative excitation of the $3P$ atoms using a laser detuned to the blue. If a large enough fraction of the atoms in the laser beam undergoes such a transition, this effect might lead to measurable temperature variations in the vapor, providing a new method of laser heating or cooling.

[We wish to point out that cooling cannot be observed in the excited state at least after one absorption process: If the laser is tuned to the red side of the line, only fast atoms can be excited from the ground state, and are slowed down, resulting in a velocity distribution which can be shown to be identical to the Maxwellian one.[10] In this case, *the ground state* is cooled.]

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