Temperature dependence of blackbody-radiation-induced transfer among highly excited states of sodium

William P. Spencer, A. Ganesh Vaidyanathan, and Daniel Kleppner
Research Laboratory of Electronics and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

Theodore W. Ducas
Department of Physics, Wellesley College, Wellesley, Massachusetts 02181
(Received 10 August 1981)

The rate of blackbody-radiation-induced transfer from the 19*s* state of sodium to the 18*p* and 19*p* states has been measured over the temperature range 6 to 210 K. The rate, which ranges from 2×10^3 to $20\times10^3\,s^{-1}$, is sensitive to several transfer coefficients and spontaneous transition rates as well as to the precise form of the radiative distribution. The results are in good agreement with theory.

I. INTRODUCTION

Blackbody radiation can be an important agent for population transfer among excited atoms. Although the transfer rate is normally small between low-lying states, radiative transfer can be large in Rydberg atoms due to the large dipole matrix elements connecting adjacent states and because the photon occupation number is often high for transitions between Rydberg states at room temperature. The ability to measure blackbody transfer is important to the understanding of the systematics of excited atoms in thermal-radiation fields, particularly for differentiating thermal-radiative transfer from transfer mechanisms such as collisions, photoionization, and other radiative processes. In addition, the ability to control blackbody-radiative transfer is essential to the application of Rydberg atoms to millimeter wave detection.[1,2]

Initial observations of the effects of blackbody radiation on Rydberg atoms were reported by Gallagher and Cooke who measured lifetime shortening and observed transfer among excited states of sodium.[3] At about the same time Beiting *et al.* independently reported observations of blackbody-radiation-induced transfer and photoionization.[4] Other effects of blackbody radiation include stimulation of superradient emission[5,6] and radiative energy-level shifts.[7] Temperature dependent radiative transfer has been observed by Figger *et al.*[2] Using a chamber cooled to 14 K, they measured transfer induced by radiation from an external source whose temperature was varied from 260 to 800 K. Koch *et al.*[8] observed effects due to transfer within a tube when the temperature was varied from 300 to 700 K. Recently Hildebrandt *et al.* reported a measurement of several transfer rates in Xe at a temperature of 90 K.[9]

In this paper, we present results on radiative transfer between the 19*s* state of sodium and the adjacent *p* states as a function of temperature over the range 6 to 210 K. In the low-temperature range of our measurements the effects of the blackbody radiation are almost completely removed, and the remaining transfer is due to spontaneous emission. We compare the measurements with our calculated results, and find good agreement.

II. THEORY

The fundamental theory governing blackbody-induced transfer is the Einstein theory of radiation.[10] The transfer rate including absorption and stimulated emission from a level i to a level j is given by

$$\gamma_{ij}=A_{ij}(\bar{n}+\theta_{ij}), \tag{1}$$

where

$$\theta_{ij}=\begin{cases}1, & E_i>E_j\\ 0, & E_i<E_j\end{cases}. \tag{2}$$

A_{ij} is the Einstein A coefficient for the transition from level i to level j and $\bar{n}$ is the average number of photons per mode at temperature T and at the transition frequency ν.

$$\bar{n}=\frac{1}{\exp\left[\frac{h\nu}{kT}\right]-1}. \tag{3}$$

The total decay rate is obtained by summing over all possible decay channels

$$\Gamma_i=\sum_{j=1}^{\infty}\gamma_{ij}. \tag{4}$$

The population in level i is governed by a rate equation for transfer in and out of the level

$$\frac{dN_i(t)}{dt}=-\sum_{j=1}^{\infty}[\gamma_{ij}N_i(t)-\gamma_{ji}N_j(t)]. \tag{5}$$

Numerical solutions to the set of rate equations is straightforward. In practice, many of the transfer rates are small and can be neglected.

Theoretical values for the A coefficients used in this paper were calculated using a numerical method described by Zimmerman *et al.*[11] The method has been shown to produce reliable matrix elements both for the calculation of the Stark effect of Rydberg atoms and for the calculation of lifetimes of Rydberg states.[12] In addition, blackbody transfer rates computed here agree well with those calculated recently by Farley and Wing.[13]

III. EXPERIMENTAL METHOD

The transfer measurements were made by initially populating the 19s state in sodium and monitoring the population as a function of time in the 19s, 18p, and 19p states over the range of temperature 6 to 210 K. These transitions were chosen because they provide wide variations in $\bar{n}$ over the available temperature range. Values for $\bar{n}$ are tabulated in Table I at several representative temperatures.

Major features of the apparatus have been described elsewhere.[12] An atomic beam of sodium is excited by pulsed dye lasers, and time resolved field ionization is used to monitor the evolution of the excited-state population. By varying the delay time to the ionizing pulse the populations in individual states are determined as a function of delay time. The signal is the sum of contributions from the initially populated state and the states which received the blackbody transfer. Because of the large difference between the s and p state spontaneous lifetimes, the resulting decay curve provided a sensitive characterization of the blackbody transfer. Measurements are repeated as the temperature is decreased from 210 to 6 K, and the data are fit to solutions of Eq. (5) describing the transfer process.

TABLE I. Values of $\bar{n}$, the average number of photons per mode in the Planck distribution, for 20-cm^{-1} radiation.

Temperature (K)	$\bar{n}$
6	0.008
20	0.31
50	1.3
210	6.8

The atoms are excited between two plates in a chamber attached to a helium cryostat (see Fig. 1). The plates are thermally connected to the helium dewar, and are surrounded by two shields also connected to the dewar. The inner shield provides electrical shielding for the plates. Apertures through the shield for the atomic beam and the lasers are covered by a 70-μ mesh and a glass window, respectively, to block far infrared radiation from outside the chamber. The helium cooled region is surrounded by a shield attached to a liquid-nitrogen reservoir.

The temperature of the outer helium shield is monitored by a copper-constantan thermocouple and a carbon resistance thermometer. These thermometers provide reliable temperature measurements through the range 4 to 300 K. Separate calibration runs were taken to assure that the field

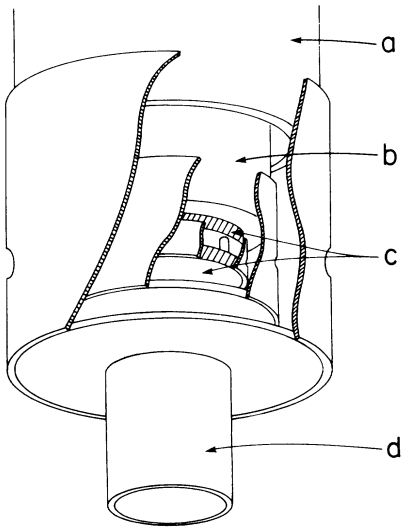

FIG. 1. Cooled containers surrounding excitation region for Rydberg atoms: (a) liquid-nitrogen reservoir, (b) liquid-helium reservoir, (c) field plates, and (d) electron multiplier housing.

plates maintain thermal equilibrium with the outer helium shield.

The sodium atoms are excited by two pulsed dye lasers whose beams counter propagate with the atomic beam. The atoms are ionized after a time delay by applying a pulsed negative voltage to the upper field plate. The delay ranges from 2 to 32 μs. The electrons emerge through small-holes in the lower field plate and pass to an electron multiplier located below. The electron multiplier is thermally connected to the liquid-nitrogen shield and is isolated from the interaction region by a radiation-blocking grid attached to the field plates. The voltage pulse is applied slowly so that the final states can be determined by time resolving the electron multiplier signal.

IV. RADIATION DISTRIBUTION

An important consideration in this experiment is how accurately the radiation distribution within the cooled chamber resembles a blackbody distribution at the wall temperature. In particular, we must consider the effect of radiation leakage through the apertures in the wall. By requiring that the flux of electromagnetic energy be in equilibrium, and assuming that the wall emissivity is small, one can obtain the following approximate formula for the radiation distribution within the chamber;

$$R_{\rm in}(\nu)=\frac{\rho(\nu,T_w)+R_{\rm ex}(\nu)a/\epsilon}{(1+a/\epsilon)}. \tag{6}$$

$\rho(\nu,T_\omega)$ denotes the Planck distribution for the wall temperature, a is the ratio of the effective area of the apertures to the area of the walls, and ϵ is the spectral emissivity of the walls. Because the apertures are covered by grids and windows whose transmission is frequency dependent, a must be treated as a function of frequency. $R_{\rm ex}(\nu)$ represents the distribution external to the helium shields, which is a mixture of Planck distributions at 6, 78, and 300 K. Using the above formula, one can estimate the relative fraction of each Planck distribution which contributes to the radiation within the interaction region. For our apparatus, within the frequency range of interest ($15\ {\rm cm}^{-1} < \nu < 60\ {\rm cm}^{-1}$), the radiation is dominated by the wall temperature Planck distribution. Assuming the grids and windows on the aperatures block no radiation and $\epsilon=0.02$, the upper limit of contributions from the 78-K distribution is 4%, and the upper limit from the 300-K distribution is 0.4%.

Another possible problem is depletion of ambient radiation within the cooled cavity due to absorption by the atoms. To estimate this effect the radiation distribution must be viewed dynamically, with the walls as the source of radiation and the atoms as a sink of radiation. If the atoms absorb at a greater rate than the walls can provide radiation, then the cavity will become supercooled at the absorption frequency. The condition for this to occur is independent of temperature since emission from the wall and absorption by the atoms are both proportional to $\bar{n}$. By comparing the absorption rate with the emission rate, one can arrive at the following condition for supercooling,

$$\frac{\epsilon\delta A_W}{N_0 A_{ij}\pi\lambda^2}<1. \tag{7}$$

A_W is the area of the walls, δ is the absorption frequency width of the atom, and N_0 is the number of excited atoms. For the parameters in our system, the quantity on the left is greater than 10^2 and this process does not occur. However in principle, this supercooling could be observed. The converse of this effect can also occur. The atoms can radiate faster than the walls can absorb, resulting in radiation trapped within the interaction cavity.

Another effect, alteration of the blackbody distribution due to the mode structure within the cooled cavity, has been considered and is expected to have little effect. The plate separation is 1 cm while the transition wavelength from the 19s to the

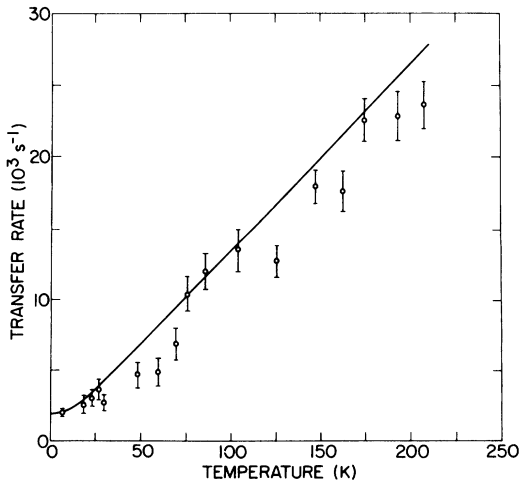

FIG. 2. Experimental and theoretical transfer rate vs temperature. The solid line is calculated with no adjustable parameters.

adjacent p states is about 0.05 cm. For these values, the region between the field plates has a mode density close to that of free space.

VI. ANALYSIS AND RESULTS

The data were analyzed by doing a least-squares fit to the solution of Eq. (5). Since we simultaneously collected the signals from the 19s, 18p, and 19p states, the solution consists primarily of the sum of an exponential decay from the s level and two curves describing transfer to the p levels. The lifetime of the 19s level used in the fit was experimentally determined from the data at 6 K to be 7.45(15) μs. Theoretical values were used for the p state lifetimes and the transfer rates to levels other than the 18p and 19p. Thus, the only free parameters in the fit were the two radiative transfer rates $\gamma_{19s,19p}$ and $\gamma_{19s,18p}$, and the scale size of the signal. More than 80% of the transfer was to the 18p and 19p states and the results were found to be insensitive to the transfer rates for which theoretical values were used.

The experimental results are shown in Fig. 2. The nonzero transfer occuring at 6 K is due to the spontaneous decay from the 19s to the 18p. We measured this decay rate to be 2040(250) s^{-1} which is in good agreement with the theoretical value of 1953 s^{-1}. The theoretical transfer curve also shown. This is given by

$$R=\gamma_{19s,18p}+\gamma_{19s,19p}. \tag{8}$$

The theoretical curve contains no adjustable parameters and is in reasonable agreement with the data. As a further check on the consistency, we fit the data to Eq. (8) to obtain experimental values for the A coefficients. The results are shown in Table II.

TABLE II. Results of fit of experimental data to Einstein A coefficients describing transfer. The calculated values are also given.

	A_{expt} (s^{-1})	A_{theor} (s^{-1})
$19s \rightarrow 19p$	1460(430)	1848
$19s \rightarrow 18p$	1950(470)	1953

Although experiment and theory are in general agreement, the experimental results for the transfer rate appear to be systematically less than the theoretical values by an amount comparable with the statistical error of each point. If the 19s lifetime were 2% shorter the fit would noticeably improve; a 4% decrease would remove any suggestion of a systematic error. We believe that the lifetime is reliable to 2%,[12] however, and that the systematic discrepancy, if it exists, probably has its origin elsewhere. We have considered other sources of error such as bias in the curve fitting algorithm, motional effects of the beam, and thermometer calibration errors, but these all appear negligible. When the data are inverted to yield values for the A coefficient, however, as in Table II, there is no evidence of a systematic error. Thus, although the possibility of a small systematic error cannot be excluded, the evidence for it is not very strong.

ACKNOWLEDGMENTS

We are grateful to Richard Cline for several useful suggestions on low-temperature techniques. This work was supported by the Office of Naval Research under Contract No. N0014-7900813 and the Joint Services Electronics Program under Contract No. DAAG29-80-C-0104.

[1]T. W. Ducas, W. P. Spencer, A. G. Vaidyanathan, W. H. Hamilton, and D. Kleppner, Appl. Phys. Lett. 35, 382 (1979).
[2]H. Figger, G. Leuchs, R. Straubinger, and H. Walther, Opt. Commun. 33, 37 (1980).
[3]T. F. Gallagher and W. E. Cooke, Phys. Rev. Lett. 42, 835 (1979).
[4]E. J. Beiting, G. F. Hildebrandt, F. G. Kellert, G. W. Foltz, K. A. Smith, F. B. Dunning, and R. F. Stebbings, J. Chem Phys. 70, 3551 (1979).
[5]M. Gross, P. Goy, C. Fabre, S. Haroche, and J. M. Raimond, Phys. Rev. Lett. 75A, 273 (1980).
[6]L. Moi *et al.*, Opt. Commun. 33, 47 (1980).
[7]W. E. Cooke and T. F. Gallagher, Phys. Rev. A 21, 588 (1980).
[8]P. R. Koch, H. Hieronymus, A. F. J. Van Raan, and W. Raith, Phys. Lett. 75A, 273 (1980).
[9]G. F. Hildebrandt, E. J. Beiting, C. Higgs, G. J. Hat-

ton, K. A. Smith, F. B. Dunning, and R. F. Stebbings, Phys. Rev. A 23, 2978 (1981).
[10]R. Loudon, *The Quantum Theory of Light* (Oxford University Press, London, 1957).
[11]M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. A 20, 2251 (1979).
[12]W. P. Spencer, A. G. Vaidyanathan, T. W. Ducas, and D. Kleppner, Phys. Rev. A 24, 2513 (1981).
[13]J. W. Farley and W. H. Wing, Phys. Rev. A 23, 2397 (1981).