

Direct measurement of diffusion coefficients of cesium $6P_{3/2}$ and $6P_{1/2}$ excited-state atoms in neon gas

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Analysis of the relative angular response of the degenerate four-wave mixing signal in alkali-metal atoms mixed with a buffer gas allows direct determination of the excited-state diffusion coefficients. The measured diffusion coefficients D at 96 °C and 760 torr for the cesium $6P_{1/2}$ and $6P_{3/2}$ state atoms in neon gas are $0.348(4) \text{ cm}^2 \text{ s}^{-1}$ and $0.281(8) \text{ cm}^2 \text{ s}^{-1}$, respectively. [S1050-2947(96)10606-5]

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The diffusion of excited alkali-metal atoms in inert buffer gases attracts interest because an excited atom can exhibit a different diffusion cross section as compared to an atom in the ground state, due to differences in interatomic potentials. Previous measurements of the diffusion of short-lived excited states have examined the light transmission of a probe beam with and without an overlapping, strong pump beam [1]. From the ratio of these two measurements, the relative change of the diffusion cross section between the ground and excited states is calculated. Recently, we have shown that it is possible to measure the excited-state diffusion coefficient for potassium atoms in xenon gas directly, using degenerate four-wave mixing in the phase-conjugate configuration [2]. The relative angular dependence of the degenerate four-wave mixing signal depends on the excited-state diffusion, and is independent of the ground-state diffusion. This method does not depend on any higher-order diffusion modes or on time-dependent analysis. Because of rapid excited-state mixing of the potassium $4P_{1/2}$ and $4P_{3/2}$ states by xenon gas, only an average diffusion coefficient of these excited states could be determined. In this paper, we report results of separately measuring the diffusion coefficients of the cesium $6P_{3/2}$ and $6P_{1/2}$ states in neon gas. To our knowledge, this is the first time that individual diffusion coefficients of alkali-metal atoms in excited states have been directly measured.

For a better understanding of the technique, we briefly review the theory that describes the experiment [2,3]. The degenerate four-wave mixing (DFWM) signal is strongly affected by atomic motion in both homogeneously broadened and inhomogeneously broadened atomic vapors [4]. In DFWM using the phase-conjugate configuration, as shown in Fig. 1, the effect of atomic motion manifests itself in the dependence of the DFWM signal on the angle between the probe and the forward pump beams [2]. At large buffer-gas densities, an atom can experience many velocity-randomizing collisions while in the excited state; therefore, the atomic motion should be characterized as diffusion rather than “free motion.” For fields below the saturation intensity

and in the steady state, we can use small-field perturbation expansion to find that the nonlinear polarization that generates the DFWM signal is proportional to the population difference between the ground and excited states, which we label as 1 and 2, respectively. This population difference can be written as [2,3]

$$\rho_2^{(2)} - \rho_1^{(2)} \propto \frac{1}{A_{21} + D_2 k_G^2} \left(1 + \frac{D_2}{D_1} \right), \quad (1)$$

where $\rho_2^{(2)} - \rho_1^{(2)}$ is the population difference with the correct phase and spatial frequency for DFWM, A_{21} is the excited-state decay rate, D_2 and D_1 are the excited- and ground-state diffusion coefficients, respectively, and $k_G = \lambda/[2 \sin(\Theta/2)]$ is the grating wave vector formed by the forward pump and probe fields (Fig. 1). The relative angular dependence arises from k_G and depends only on the excited-state diffusion coefficient D_2 and the decay rate A_{21} . The ground-state diffusion coefficient enters only in a constant multiplicative factor that sets the overall magnitude of the induced nonlinear polarization. There is also a similar equation that describes the grating formed by the backward pump and probe fields, but this term is much smaller, and its contribution has been included in the analysis.

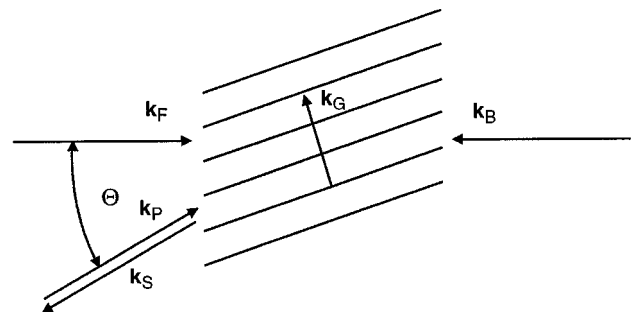


FIG. 1. DFWM using the phase-conjugating configuration. It shows the grating written by the forward pump and probe beams, and the signal beam that results when the backward pump scatters from the grating. There is also a similar grating formed by the probe and backward pump beams.

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In general, the two-level model outlined above needs to be modified due to fine-structure-mixing collisions between the $6P_{3/2}$ and $6P_{1/2}$ states. For cesium, at the neon pressures used in our experiments, the collisional mixing rate between the $6P_{1/2}$ and $6P_{3/2}$ states is much smaller than the excited-state decay rate, so that the above two-level analysis remains valid [5,6]. For our conditions, we expect to measure distinct diffusion coefficients for excitation to each P state.

Our experimental setup was similar to that described elsewhere [7]. Cesium vapor was contained in Pyrex cells with window separations of 1 mm. The cells were prepared by evacuating them to below 10^{-6} torr, distilling in a small amount of natural cesium metal, introducing the desired pressure of Ne, and finally sealing the cell. For this experiment, cells containing 1150, 1850, and 2300 torr Ne at room temperature were prepared [4]. The exact buffer-gas pressures in the vapor cells were determined by measuring the line broadening of the atomic absorption due to the collisions by the buffer gas. The cells were heated to a temperature of 96°C , at which the linear intensity attenuation length was greater than the cell length for both $6S_{1/2}$ - $6P_{1/2}$ and $6S_{1/2}$ - $6P_{3/2}$ transitions of every cell. A frequency-stabilized Ti:sapphire laser was used to generate a probe, a forward pump, and a counterpropagating backward pump beam. All three input beams were vertically polarized, and the generated signal beam had the same polarization. The forward pump-probe angle was varied from 20 to 400 mrad. The forward pump beam was chopped, and the signal was measured with a lock-in amplifier.

All three input beams had nearly the same intensity (2 W cm^{-2}). The total laser intensity was less than the two-level saturation intensity for the homogeneously broadened $6S_{1/2}$ - $6P_{1/2}$ and $6S_{1/2}$ - $6P_{3/2}$ transitions ($\sim 20\text{ W cm}^{-2}$). This ensured that the use of the third-order perturbation expansion for the DFWM signal was valid. For these pressures we estimated that there would be in excess of 400 velocity randomizing collisions during the excited-state lifetime, and our diffusion model should be valid. For all cells, the homogeneous broadening [8] (9.8 MHz/torr) was larger than the Cs ground-state hyperfine splitting (9.19 GHz), as well as the Doppler width (450 MHz). The laser frequency was tuned to the center of the pressure-shifted resonance, and the DFWM signal was measured as a function of the angle between the two forward-going beams.

Figure 2 shows the data taken at the $6S_{1/2}$ - $6P_{1/2}$ transition. The circles and squares show the measured DFWM signal as a function of the forward pump-probe angle at the conditions described above. The lines in Fig. 2 are least-squares fits to the data using Eq. (1). The fitting procedure had two free parameters, the excited-state diffusion coefficient and the zero-angle signal. Similar data were taken using the $6S_{1/2}$ - $6P_{3/2}$ transition. For these fits we used excited-state lifetimes of 34.9 and 30.5 ns for the $6P_{1/2}$ and $6P_{3/2}$ states [9], respectively, since there was little excited-state quenching. From analysis of the DFWM signal angular dependence for each cell, we were able to determine the diffusion coefficient at each pressure at the same temperature.

Figure 3 shows the results for the diffusion coefficient plotted versus the reciprocal of the neon pressure at 96°C . These data clearly show a difference in diffusion coefficient, depending on which transition is utilized. At a constant tem-

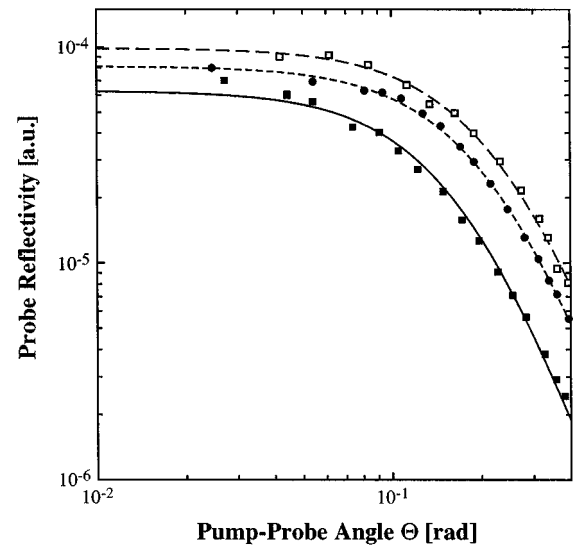


FIG. 2. Experimental DFWM signals as a function of forward pump-probe angle for the Cs $6S_{1/2}$ - $6P_{1/2}$ transition. The solid boxes, solid circles, and open boxes are measured reflectivities from cells containing 1150, 1850, and 2300 torr of neon gas, respectively. The solid, short-dashed, and long-dashed lines are least-squares fits of Eq. (1) for each cell.

perature, the diffusion coefficient is inversely proportional to the pressure, and we have performed a least-squares fit of our experimental data to determine the constant of proportionality D , which is the excited-state diffusion coefficient at 760 torr (pressure at 96°C). Our results give values for D at 96°C of $0.348(4)\text{ cm}^2\text{ s}^{-1}$ for the $6P_{1/2}$ state, and $0.281(8)\text{ cm}^2\text{ s}^{-1}$ for the $6P_{3/2}$ state. Hamel *et al.* [5] have calculated diffusion cross sections for the $6P_{1/2}$ and $6P_{3/2}$ states of

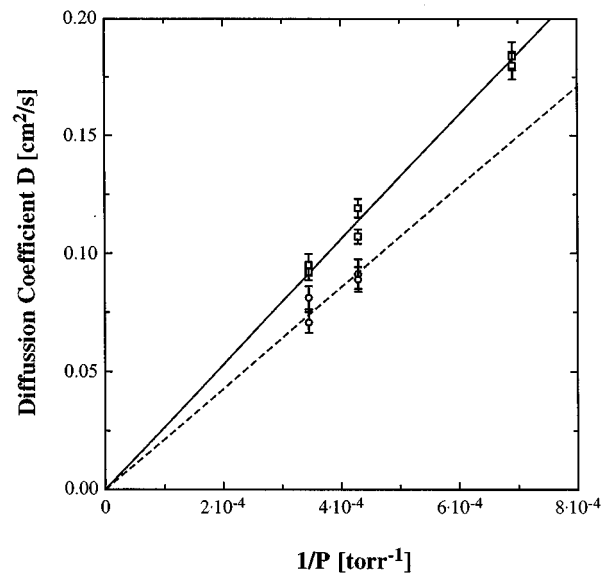


FIG. 3. Plot of measured D at 96°C vs $1/P$ in torr (pressures at 96°C). Squares and circles with error bars show experimental results from data similar to Fig. 2, for the $6S_{1/2}$ - $6P_{1/2}$ and $6S_{1/2}$ - $6P_{3/2}$ transitions, respectively. The lines are least-squares fits for each transition, and give the diffusion coefficients stated in the text.

cesium in neon at 47 °C to be $\sigma_{P_{1/2}}=36.4 \text{ \AA}^2$ and $\sigma_{P_{3/2}}=68.5 \text{ \AA}^2$, respectively. From these values, we calculate that the diffusion coefficients at 96 °C are $D_{P_{1/2}}=0.295 \text{ cm}^2 \text{ s}^{-1}$ and $D_{P_{3/2}}=0.157 \text{ cm}^2 \text{ s}^{-1}$. Our experimental data confirm that the $6P_{3/2}$ state has a smaller diffusion coefficient than the $6P_{1/2}$. However, the agreement between our measurements and the theoretical values is poor. The theoretical potentials used in calculating the cross section are known to be not the best for all alkali-metal noble-gas pairs [5]. Therefore, the lack of agreement between the measured and calculated diffusion coefficients indicates that the potentials used by Hamel *et al.* [5] are not appropriate. The experimental values presented here should prove useful in evaluating refined interaction potentials.

In conclusion, we have measured the diffusion coeffi-

cients for the $6P_{1/2}$ and $6P_{3/2}$ state cesium atoms in neon gas. These diffusion coefficients were determined from the relative angular dependence of DFWM. The above-presented data were obtained in the steady state, and did not rely on any time-dependent signal decay to extract the diffusion parameter. This procedure can be extended to measure the excited-state diffusion coefficients for other atoms in an inert gas. The increased flat angular response observed using a buffer gas is also important for optical information processing [10,11], since this will increase the number of pixels in the images that can be processed.

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