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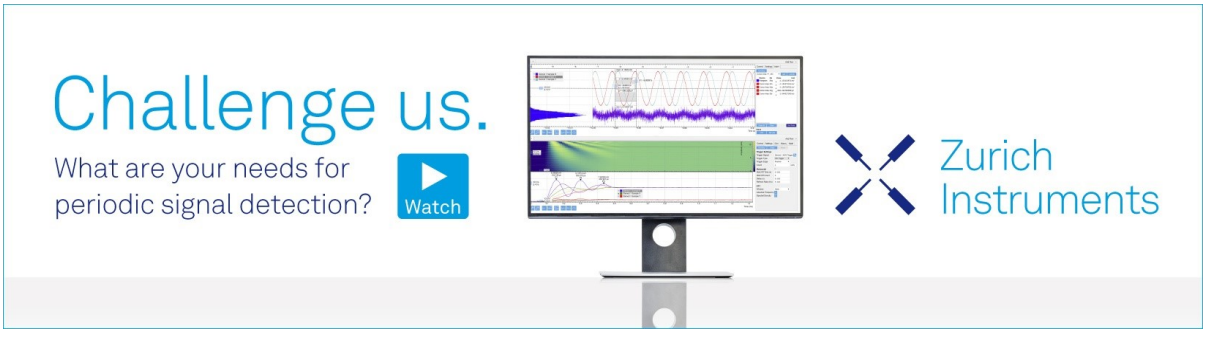

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Hyperfine and Zeeman quantum beats of the Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) transition, and the decay of coherence

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Quantum beats of the Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) transition and their change with the magnetic field were observed in pure Na vapor and Na + He gas by using a picosecond laser system. The energies of the hyperfine and Zeeman sublevels of the $3\,^2P_{3/2}$ and $3\,^2S_{1/2}$ states were calculated by using the reported hyperfine constants and g factors, and the theoretical curve of the quantum beats was calculated. The results were in good agreement with the observed profile of the quantum beats in phase of modulation, but the difference of the amplitudes increased with time. By comparing the amplitude of the theoretical curve of the quantum beats with that of the experimental curve, we determined the relaxation time of coherence. The decay of amplitude of the modulation was found to become faster as the pressure of the foreign gas (He) was increased. The decay of amplitude, i.e., the decay of coherence arises from the dephasing, which occurs from the fluctuation of the level energies induced by the interatomic interaction.

I. INTRODUCTION

Quantum beat spectroscopy is a very useful Doppler-free high resolution spectroscopy to investigate interacting levels with small energy splittings and has been successfully applied.[1–6] Because of the hyperfine splitting of the $n\,^2P_{3/2}$ state of alkali metal atoms, the quantum beats can be observed in the zero magnetic field.[7–9] The hyperfine quantum beats of the Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) transition were observed previously by the technique of the fast atomic beam combined with the laser excitation[10]; the crossing of an atomic beam with a well collimated continuous laser beam led to a pulse excitation during the time interval of the passing. The quantum beats of the hyperfine levels of the Na $3\,^2P_{3/2}$ state were also observed in photoionization.[11]

By using a pulsed laser of 10 ps duration, we observed the quantum beats of the resonance fluorescence of the Na D_2 ($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) line both in the absence and in the presence of an external magnetic field. The magnetic sublevels split in the presence of the magnetic field, and the time profile of the quantum beats changed with the magnetic field. The time profile is determined by the energy splittings of the $3\,^2P_{3/2}$ state and the transition moments between the sublevels of the $3\,^2P_{3/2}$ and $3\,^2S_{1/2}$ states. We shall analyze the time profile of the observed quantum beats by using the hyperfine constants and the g factors of Na atom previously determined by various methods,[12–15] and evaluate the decay of the coherence. We shall report the first observation of the relaxation of the coherence, which arises from the dephasing, induced by collision.

II. EXPERIMENTAL

A schematic diagram of the apparatus is shown in Fig. 1. A small amount of sodium metal was sealed *in vacuo* in a Pyrex rectangular cell with a side arm reservoir ($10\times10\times30$ mm), which was baked at 800 K for several hours and the residual pressure was lower than 1×10^{-6} Torr. Foreign gas He was then added to study the collisional effects. In order to prevent condensation of sodium vapor on the cell window, the side arm was maintained at a temperature about 10 K below that of the cell window. We used the picosecond pulsed laser and single photon counting system which has been previously described in detail.[16] We excited the Na D_2 line by a linearly polarized pulsed laser (duration 10 ps, bandwidth 6 cm^{-1}). The fluorescence was dispersed by a monochromator (Nikon P250) through a polarization analyzer and a polarization scrambler and detected the D_2 line by a microchannel plate photomultiplier (Hamamatsu R1564U-01). Time-resolved fluorescence intensity was

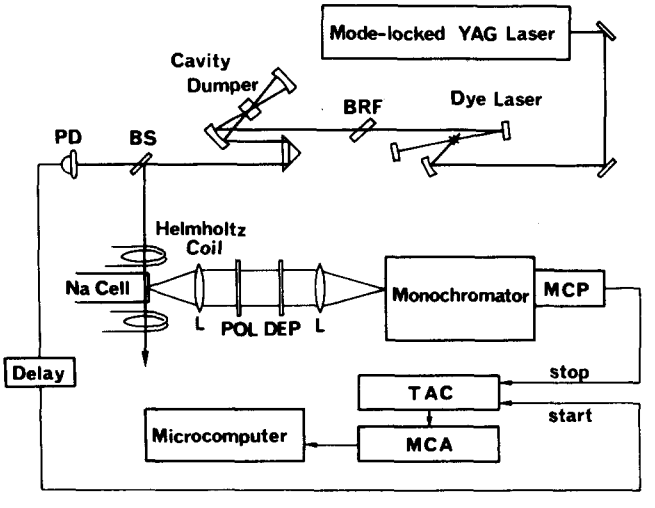

FIG. 1. Schematic diagram of the apparatus. TAC: time-to-amplitude converter; MCA: multichannel pulse height analyzer; MCP: microchannel plate photomultiplier; PD: photodiode; BS: beam splitter; BRF: birefringent filter; POL: polarization analyzer; DEP: polarization scrambler.

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measured by a time-to-amplitude converter and a pulse height analyzer. We used a Helmholtz coil to observe the Zeeman quantum beats. The magnetic field was applied parallel to the traveling direction of the laser light.

III. RESULTS AND DISCUSSION

We observed the time-resolved intensity of resonance fluorescence of the Na D_2 ($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) line after the excitation by a pulsed laser both in the presence and in the absence of a magnetic field. We found prominent quantum beats when the polarization of the analyzer was parallel to the electric vector of the laser light (σ component; perpendicular to the magnetic field). The results are shown in Fig. 2. The frequency of the intensity modulation increased as the magnetic field was increased.

The decay of the fluorescence intensity can be decomposed into two components; one is a monotonous exponential decay (A component) and another is oscillating beats (B component). One of the results at the magnetic field of 128 G is shown in Fig. 3. The lifetime of the A component, which is equal to the radiative lifetime, was 16.6 ns at the pressure less than 10^{-4} Torr. This is in agreement with the reported lifetime 16.3 ± 0.4 ns.[17] The time profile of the B component shows a superposition of several sequences of quantum beats with different frequencies. We shall analyze this profile.

The Hamiltonian for the hyperfine structure of an atom in the magnetic field H is given by,[4]

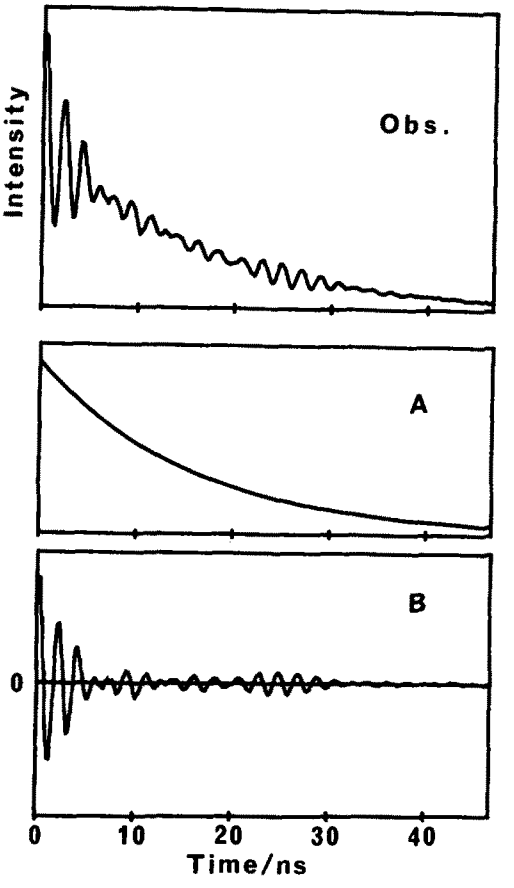

FIG. 3. Observed decay profile of the σ component of Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) resonance fluorescence at 128 G (Obs.). A and B are, respectively, the monotonous exponential decay component and the oscillating beat component. The vapor pressure of Na is 1.3×10^{-6} Torr.

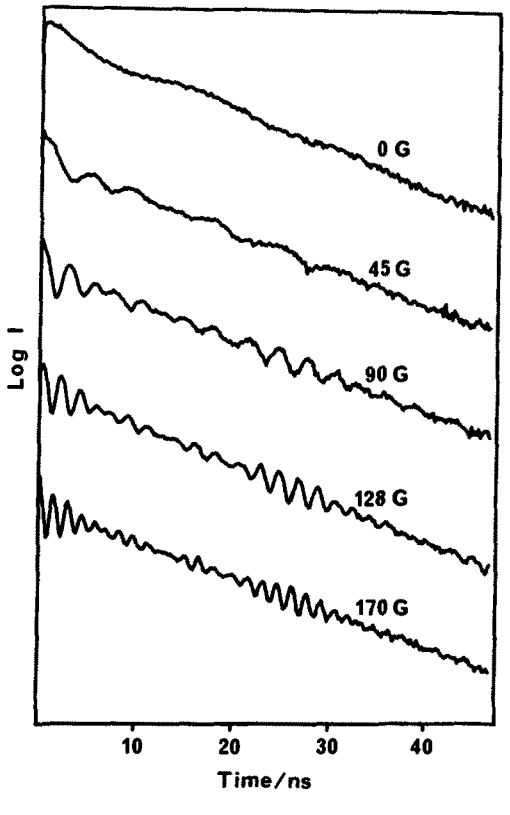

FIG. 2. Observed decay profiles (log I, where I is the intensity) of the σ component (polarized parallel to the electric vector of laser light and perpendicular to an applied magnetic field) of Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) resonance fluorescence in various magnetic fields. The vapor pressure of Na is 1.3×10^{-6} Torr.

$$H_{\mathrm{HFS}} = A_J \mathbf{I}\cdot\mathbf{J} + B_J[3(\mathbf{I}\cdot\mathbf{J})^2 + 3(\mathbf{I}\cdot\mathbf{J})/2 - I(I+1)J(J+1)]/2I(I+1)J(J+1) + g_J\mu_B\mathbf{J}\cdot\mathbf{H} - g_I\mu_B\mathbf{I}\cdot\mathbf{H}, \quad (1)$$

where I and J are the nuclear spin and total electronic angular momenta, respectively. The first term represents the magnetic dipole interaction and the second term represents the electric quadrupole interaction, and A_J and B_J are the constants. The third and fourth terms represent, respectively, the electronic Zeeman interaction and the nuclear Zeeman interaction, g_J and g_I are the g factors of the electron and nucleus, respectively, and μ_B is the Bohr magneton. These hyperfine constants and g factors have been determined by extensive spectroscopic studies. We used the values obtained by the double resonance and beam experiments[4,12]: $A_J = 18.65$ MHz, $B_J = 2.82$ MHz, $g_J = 1.3344$ and $g_I = -0.0008$ for the $3\,^2P_{3/2}$ state, and $A_J = 885.82$ MHz, $B_J = \sim 0$ MHz, $g_J = 2.0023$ and $g_I = -0.0008$ for the $3\,^2S_{1/2}$. We then constructed the energy matrix with the basis set of $|J\,M_J\,I\,M_I\rangle$ and obtained the eigenvalues and eigenfunctions. The resultant energy levels are shown as a function of the magnetic field strength in Fig. 4.

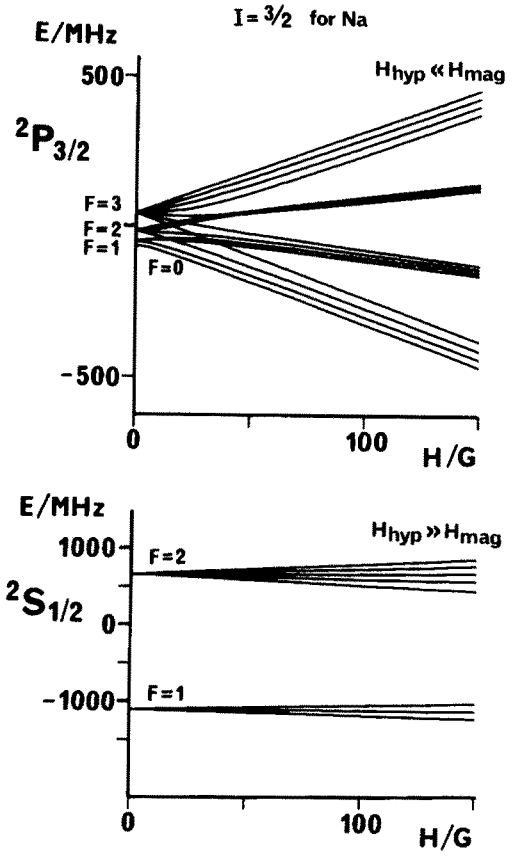

FIG. 4. Energy level diagram of the $3\,^2P_{3/2}$ and $3\,^2S_{1/2}$ states of Na with nuclear spin 3/2 as a function of a magnetic field H.

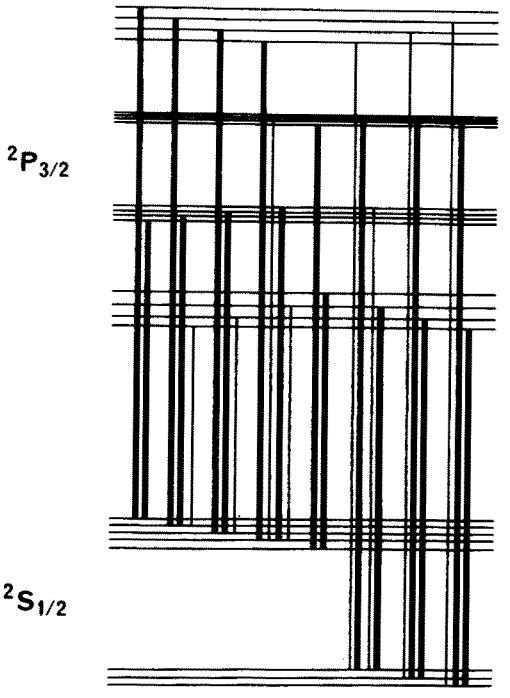

FIG. 5. Calculated transition moments of the σ component between hyperfine sublevels of the $3\,^2P_{3/2}$ and $3\,^2S_{1/2}$ states at the magnetic field of 128 G. Thick lines show large transition moments and thin lines show small ones.

The nonvanishing matrix elements of the σ-polarized transition moments between the $3\,^2P_{3/2}$ and $3\,^2S_{1/2}$ states are given by[18]

$$\langle {}^2S_{1/2} J M_J 3/2 M_I | \mu_X | {}^2P_{3/2} J+1 M_J \pm 1\, 3/2 M_I \rangle = \mp \langle {}^2S_{1/2} J \| \mu \| {}^2P_{3/2} J+1 \rangle \times \{(J \pm M_J + 1)(J \pm M_J + 2)\}^{1/2}/2, \quad (2)$$

where $\langle {}^2S_{1/2} J \| \mu \| {}^2P_{3/2} J+1 \rangle$ is the reduced matrix element. The transition moments between the eigenstates at 128 G are calculated, and the results are schematically shown in Fig.5.

When atoms in the initial state $|i\rangle$ are coherently excited to the levels $|m\rangle$ by an optical pulse, the atoms are described by a coherent superposition of the excited states,[2,4]

$$\phi(t) = \sum_m \langle m|\mu|i\rangle |m\rangle \times \exp[-i(E_m/\hbar)t]\exp[-(\Gamma/2)t], \quad (3)$$

where μ is the dipole moment, E_m is the energy of the level $|m\rangle$, and Γ is the decay constant. When atoms in this superposition state decay to a common lower level $|f\rangle$, the intensity of the emitted light is given by

$$I(t) \propto |\langle f|\mu|\phi(t)\rangle|^2. \quad (4)$$

The intensity is modulated at the frequency corresponding to the energy difference between the excited levels $|m\rangle$. We used a pulsed laser of the band width 6 cm^{-1} = 180 GHz, and it was sufficiently broad compared with the Zeeman splitting of the Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) transition at $H = 128$ G. Hence, all the allowed transitions, which are shown in Fig. 5, occurred simultaneously. The expression for the time-dependent emitted light intensity is given by

$$\begin{aligned} I(t) &\propto \sum_{if}\sum \Big| \sum_m \langle f|\mu|m\rangle\langle m|\mu|i\rangle \times \exp[-iE_m t/\hbar]\exp[-(\Gamma/2)t] \Big|^2 \\ &= \sum_{ifm}\sum\sum |\langle f|\mu|m\rangle|^2 |\langle m|\mu|i\rangle|^2 \exp(-\Gamma t) \\ &\quad + \sum_{\substack{ifmm' \\ (m\neq m')}}\sum\sum\sum \langle f|\mu|m\rangle\langle m|\mu|i\rangle\langle f|\mu|m'\rangle\langle m'|\mu|i\rangle \times \exp[-i(E_m - E_{m'})t/\hbar]\exp(-\Gamma t) \\ &= \sum_{ifm}\sum\sum |\langle f|\mu|m\rangle|^2 |\langle m|\mu|i\rangle|^2 \exp(-\Gamma t) \\ &\quad + 2\sum_{\substack{ifmm' \\ (m>m')}}\sum\sum\sum \langle f|\mu|m\rangle\langle m|\mu|i\rangle\langle f|\mu|m'\rangle\langle m'|\mu|i\rangle \times \cos[(E_m - E_{m'})t/\hbar]\exp(-\Gamma t). \end{aligned} \quad (5)$$

The first term corresponds to the exponential decay (A component) and the second term corresponds to the quantum beats (B component). The time profile of the quantum beats depends on energies of the hyperfine and Zeeman sublevels and the transition moments between the levels.

By using the calculated energies for all the hyperfine and Zeeman sublevels of the $3\,^2P_{3/2}$ and $3\,^2S_{1/2}$ states and the transition moments between them, we calculated the intensity modulation, i.e., the second term of Eq. (5) multiplied by $\exp(+\Gamma t)$. The lower trace of Fig. 6 (Cal.) shows the calculated result for the intensity modulation at 128 G. The upper trace of Fig. 6 (Obs.) shows the observed intensity modulation, which is the B component in Fig. 3 multiplied by $\exp(+\Gamma t)$ with the radiative lifetime $1/\Gamma = 16.6$ ns. Observed and calculated intensity modulations are in good agreement in phase. We could reproduce the observed beat profiles at any other magnetic fields by similar calculations. Thus, the frequency of quantum beats in the resonance fluorescence of Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) transition and its change with the magnetic field can be understood on the basis of the energy levels which are calculated from the reported hyperfine constants and g factors. However, the amplitude of the observed modulation (Obs.) gradually decays and the difference from the calculated amplitude (Cal.) increases with time (see Fig. 6). The amplitude of the modulation was found to decay rapidly as the vapor pressure of Na atoms was increased by raising the temperature (370 K, 8.6×10^{-8} Torr ~ 470 K, 1.3×10^{-4} Torr). However, an accurate measurement of the lifetime is difficult, because the increase of the vapor pressure of Na atoms increases the self-reversal. In order to observe the collisional effects, we added the helium gas keeping the vapor pressure of Na at 1.5×10^{-7} Torr and the temperature at 375 K. The observed changes of the beat profile are shown in Fig. 7.

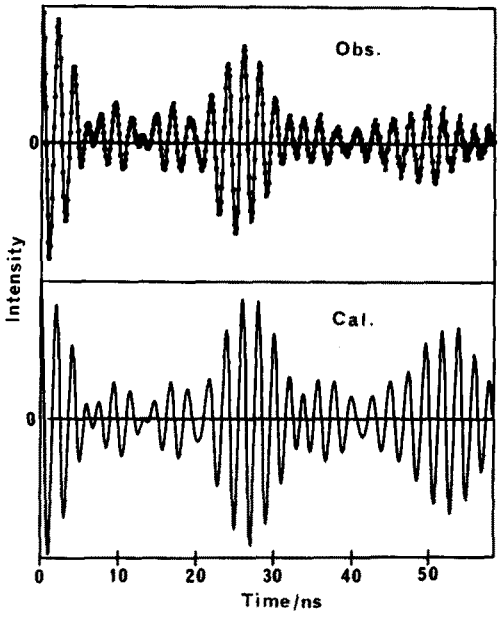

FIG. 6. The intensity modulation of quantum beats at 128 G. The lower trace (Cal.) shows the calculated values of $2\Sigma\Sigma\Sigma\Sigma_{(m>m')}$ $\langle f|\mu|m\rangle\langle m|\mu|i\rangle\langle f|\mu|m'\rangle\langle m'|\mu|i\rangle\cos[(E_m - E_{m'})t/\hbar]$ for the σ component of Na($3\,^2P_{3/2}$–$3\,^2S_{1/2}$) resonance fluorescence. The upper trace (Obs.) shows the observed intensity modulation, which is the B component in Fig. 3 multiplied by $\exp(+\Gamma t)$ where $1/\Gamma = 16.6$ ns.

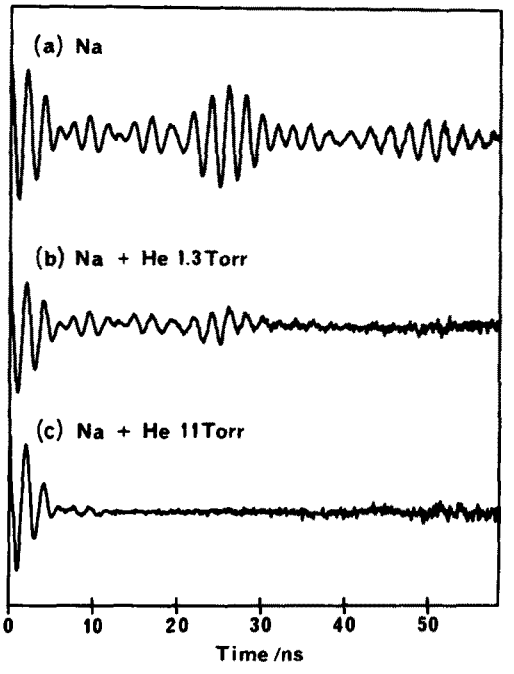

FIG. 7. The change of the intensity modulation at 128 G by adding He gas. (a) is the observed intensity modulation for pure Na vapor. (b) and (c) are the ones for Na vapor + He(1.3 Torr) and Na vapor + He(11 Torr), respectively. The vapor pressure of Na is 1.5×10^{-7} Torr in all cases.

The frequency of the quantum beats and the radiative lifetime did not change with the foreign gas pressure. However, the amplitude of the modulation decayed rapidly as the foreign gas pressure was increased. The decay of the amplitude of the modulation was approximately single exponential. The decay times were determined by the least-squares fitting, and were found to be 62 ns for (a) Na, 17 ns for (b) Na + He(1.3 Torr), and 5.3 ns for (c) Na + He(11 Torr) at 375 K. This shows that the decay of amplitude of the modulation, i.e., the decay of coherence, is more sensitive to the foreign gas pressure than the radiative decay. The decay of coherence arises from the dephasing induced by collision. The dephasing occurs from the fluctuation of the level energies induced by the interatomic interaction. While the radiative lifetime is shortened by the collisional energy transfer between the electronic states:

$$\mathrm{Na}(3\,^2P_{3/2}) + \mathrm{He} \rightarrow \mathrm{Na}(3\,^2P_{1/2}) + \mathrm{He}. \qquad (6)$$

The decay of coherence may be induced more effectively by collision than the shortening of the radiative lifetime because the energy splittings of the magnetic sublevels in the $3\,^2P_{3/2}$ state are small compared with the energy difference between the $3\,^2P_{3/2}$ and $3\,^2P_{1/2}$ states ($17.2\ \mathrm{cm}^{-1}$).

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