# Measurement of the collisional depolarization cross section of the silver $5^2P_{1/2}$ state by helium

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We have measured the cross section for depolarization of the $5^2P_{1/2}$ state of silver atoms by collisions with helium atoms using the Hanle-effect technique. We find the nuclear-spin-independent cross section to be $\sigma = 1.2 \pm 0.15 \times 10^{-16}$ cm$^2$, significantly smaller than the corresponding cross sections in rubidium and cesium. This is the first measurement of this quantity in any noble-metal atom.

## I. INTRODUCTION

Single-valence-electron atoms are a very important class of atoms due to their simple spectra and the relative ease with which they may be treated theoretically. Of all the inelastic collision processes, collisional depolarization process, i.e., mixing of various $m_J$ sublevels under the influence of a perturbing atom, is perhaps the simplest. Although it is not an easy matter to deduce interatomic potentials from the experimental cross sections, the experimental cross sections can nevertheless be used to check theoretical cross sections derived from different interatomic potentials. The experimental cross sections also contribute to our understanding of the role played by various factors, such as nuclear spin, spin-orbit interaction, etc., in these collisions. For this reason, extensive work, both theoretical and experimental, on the depolarization of alkali-metal atoms by inert gases has been done during the last two decades.[1] However, no corresponding work on noble-metal atoms (which are also single-valence-electron atoms) has been done. In this paper we present the results of our investigation of the depolarization of the $5^2P_{1/2}$ state of silver-107 by helium. We find this cross section to be very small compared to that of alkali-metal atoms. This is the first measurement of this quantity in any noble-metal atom.

If the atoms are excited by polarized radiation, the excited state $m_J$ sublevels may be populated unequally. In other words, the excited state may acquire a certain degree of electronic polarization. Owing to collisions with inert-gas atoms, the populations of different $m_J$ sublevels tend to equalize or, equivalently, the excited state tends to get depolarized. The aim of the experiments on alkali-metal atoms has been to measure cross sections for these processes. The population distribution among $m_J$ sublevels can be expressed in terms of atomic multipoles,[1,2] for example, $P_{1/2}$ population distribution can be expressed as a sum of monopole (population) and dipole (orientation) distributions. Similarly, $P_{3/2}$ population distribution can be expressed as a sum of monopole (population), dipole (orientation), quadrupole (alignment), and octupole distributions. The atomic multipolarity does not change for isotropic collisions, and different multipoles relax at different rates. In order to obtain complete information on the collision process, one needs to measure all different multipole relaxation cross sections. This is generally possible; however, octupole relaxation rate cannot be measured by using polarization of the radiation as a probe.[2]

The nuclear spin plays a very important role in these collisions.[3] Owing to hyperfine coupling, part of the electronic polarization created by the polarized exciting light is transferred to nuclear polarization. Since in most cases investigated the hyperfine precession period is much longer than the duration of the collision, the nuclear polarization is not affected by the collision. On the other hand, the electronic angular momentum is randomized by the collision. Since the nucleus can store large amounts of angular momentum, it can regenerate the electronic polarization via hyperfine coupling, making the observed depolarization cross sections appear too small.[3] These effects can be quantitatively taken into account.[3,4]

Almost all of the investigations in alkali-metal atoms have been performed on the first excited $P_{1/2}$ and $P_{3/2}$ states.[1] Several different techniques have been utilized for these investigations. These include optical pumping,[5,6] Hanle effect[7] and high-field level crossings,[8] and depolarization of the fluorescence after selective Zeeman excitation.[9] Recently, individual cross sections for $Jm_J \rightarrow J'm_J'$ mixing have been measured by selective excitation of a $Jm_J$ sublevel in a high magnetic field (up to 131 kG) and observation of fluorescence from isolated $J'm_J'$ sublevels.[10,11] These measurements give the most complete information about the collisions. Some of the noteworthy results of depolarization cross sections in alkali-metal atoms are the following: The $P_{1/2}$ depolarization cross sections are much smaller than the corresponding cross sections in the $P_{3/2}$ states of heavy-alkali atoms.[12] Small $P_{1/2}$ state cross sections are understood[1,13,14] as being due to a selection rule that requires that the mixing proceed via a virtual transition to a $P_{3/2}$ state. Also, a magnetic field dependence of the $P_{1/2}$ cross sections is found[15] which, at least partially, may be due to the mixing of $P_{1/2}$ and $P_{3/2}$ states in a strong magnetic field.

In this paper we present results of the first measurement of depolarization cross section in a different class of single-valence-electron atoms—noble-metal atoms. In particular, we have measured the disorientation cross sections for the $5^2P_{1/2}$ state of silver atoms for collisions

 

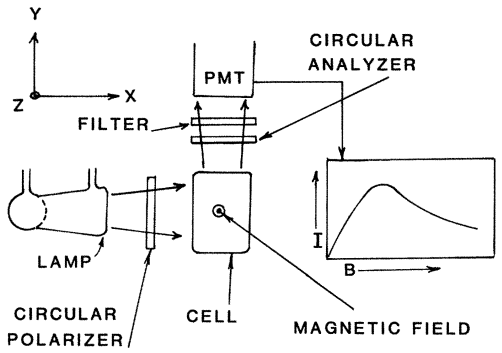


FIG. 1. Schematic illustration of the experiment. Circularly polarized silver resonance radiation from an rf discharge lamp, propagating along the $x$ axis, is absorbed by silver vapor. Fluorescent light, filtered by a 40-Å bandwidth filter, and analyzed for circular polarization is observed along the $y$ axis. When observed as a function of an external magnetic field in the $z$ direction, collision broadened Hanle-effect signal, as sketched in the box, is obtained.

with helium atoms using the Hanle-effect technique.

The experiment is shown schematically in Fig. 1. Circularly polarized 3382.9 Å ($5^2P_{1/2}\rightarrow 5^2S_{1/2}$) resonance radiation from an rf excited silver lamp is incident, let us say, along the $x$ direction on a quartz cell containing separated isotope of silver-107. The cell is heated to about 1000 K to get enough vapor pressure of silver. 3382.9 Å fluorescent radiation, after passing through a circular polarization analyzer, is detected in the $y$ direction by a photomultipler tube. A dc magnetic field is applied in the $z$ direction. The fluorescent light is observed as a function of the magnetic field. Circularly polarized exciting light produces atomic orientation in the $x$ direction, which precesses in the magnetic field. The observed fluorescent light has a magnetic field dependence as sketched in Fig. 1 (Hanle effect).[16,17] The width of the Hanle curve depends on the radiative lifetime and $g_J$ value of the excited state. When inert gas is introduced into the cell, the width of the curve increases due to collisional depolarization of the $P_{1/2}$ state. The depolarization cross section is deduced from a measurement of the broadening of the Hanle curve as a function of the inert-gas pressure.

Results of the theory of Hanle effect in the pressure of a buffer gas, including the effects of nuclear spin,[3] are given in Sec. II. The experiment is described in Sec. III and the results are discussed in Sec. IV.

## II. THEORY

The theory of the Hanle effect in the presence of depolarizing collisions is given by Bulos and Happer (BH).[3] The effects of nuclear spin are explicitly taken into account in this theory. Theoretical results of BH apply to our case exactly. Therefore, we will quote their results without rederiving them.

Consider the experimental situation outlined in Fig. 1, i.e., circularly polarized resonance radiation propagating along the positive $x$ axis excites the atoms. An external magnetic field defines the $z$ axis, and circularly polarized fluorescent radiation is detected along the positive $y$ axis. The intensity of the fluorescent light $\Delta I$ observed in a small solid angle $\Delta\Omega$ is given by (see BH)

$$\frac{\Delta I}{\Delta\Omega}=C(1+2\vec{s}\cdot\langle\vec{J}\rangle_e)\,, \tag{1}$$

where $C$ is a constant (which depends on the oscillator strength of the transition), $\vec{s}$ is the mean spin of the detected photons, and $\langle\vec{J}\rangle_e$ is the expectation value of the electronic angular momentum. For our experimental situation, Eq. (1) reduces to

$$\frac{\Delta I}{\Delta\Omega}=C(1\pm 2\langle J_y\rangle) \tag{2}$$

depending on whether right or left circularly polarized light is observed. Equation (2) predicts that the observed intensity is proportional to the transverse electronic polarization $\langle J_y\rangle$. BH have derived an expression for $\langle J_y\rangle$,

$$\langle J_y\rangle=\frac{R}{3[I]^2}\omega\left[\Gamma^2\left[3I+\frac{3}{2}\right]+\frac{2\Gamma\gamma}{[I]}(4I^2+4I+3)+\frac{6\gamma^2}{[I]}+\omega^2\left[3I+\frac{3}{2}\right]\right]\Big/\left\{\left[\left[\Gamma+\frac{2\gamma}{[I]^2}\right](\Gamma+\gamma)+\omega^2\right]^2+\frac{\gamma^2\omega^2}{[I]^2}\right\}, \tag{3}$$

where

$$[I]\equiv 2I+1\,, \tag{4}$$

$\omega$ is the Larmor frequency,

$$\omega=g_J\left[\frac{\mu_B}{\hbar}\right]\frac{1}{2I+1}B\,, \tag{5}$$

and $R$ is the mean excitation rate. In Eq. (3), $\Gamma=\tau^{-1}$ is the radiative decay rate and $\gamma$ is the *electronic* depolarization rate given by

$$\gamma=n\bar{v}\sigma\,, \tag{6}$$

where $n$ is the density of inert-gas atoms, $\bar{v}$ is the relative velocity of the collision partners, and $\sigma$ is the depolarization cross section.

In the derivation of Eq. (3) the following assumptions have been made: (1) The spectral profile of the exciting light is effectively flat over the absorption width of the atoms. (2) The external magnetic field is sufficiently small that linear Zeeman effect is a good assumption. (3) The hyperfine precession frequency is much larger than

the natural decay rate, i.e., $A \gg \Gamma/2\pi$ where $A$ is the magnetic dipole coupling constant. All of the above conditions are approximately valid in our experiment.

Equation (3) predicts that the electronic polarization $\langle J_y \rangle$ is zero at zero magnetic field, increases to a maximum value for some value of the magnetic field $B_0$, and falls gradually for still higher magnetic fields as sketched in Fig. 1. In the absence of depolarization collisions, Eq. (3) reduces to the usual Hanle-effect signal given by

$$\frac{\omega}{(\Gamma^2+\omega^2)} \times \text{const} \tag{7}$$

with a width

$$\omega_0 = \Gamma \ , \tag{8}$$

where

$$\omega_0 = g_J \left[ \frac{\mu_B}{\hbar} \right] \frac{1}{[I]} B_0 \ . \tag{9}$$

In the presence of depolarizing collisions ($\gamma \neq 0$), the effective width of the signal can be approximated by

$$\omega_0 = (\Gamma^2 + a\Gamma\gamma + b\gamma^2)^{1/2} \ , \tag{10}$$

where

$$a = \frac{2}{3[I]^2}(4I^2+4I+3) \ , \tag{11}$$

and

$$b = \frac{1}{[I]^2} \ . \tag{12}$$

An inspection of Eqs. (10)–(12) reveals that the effective width in the absence of nuclear spin ($I=0$) is

$$\omega_0 = \Gamma + \gamma \ , \tag{13}$$

as expected. However, the width is reduced in the presence of nuclear spin, as given by Eq. (10). For example, in the case of $^{107}$Ag ($I = \frac{1}{2}$), the effective width is given by

$$\omega_0 = \Gamma + \tfrac{1}{2}\gamma \ . \tag{14}$$

The physical reason for reduction in the width is the nuclear "flywheel effect." When the circularly polarized light creates electronic polarization, due to hyperfine coupling, part of the polarization is transferred to the nucleus. The nucleus, being massive, can absorb a large amount of angular momentum. During the collision, electronic polarization is destroyed. However, since the nuclear precession time ($\gtrsim 10^{-10}$ sec) is much larger than the duration of the collision ($\gtrsim 10^{-12}$ sec), the nucleus does not "feel" the effect of the collision and it remains essentially inert during collisional depolarization of the electronic angular momentum. After the collision is over, the nucleus regenerates part of the electronic angular momentum, reducing the effective depolarization rate.

## III. EXPERIMENT

Details of the experimental setup are shown schematically in Fig. 2. Resonance radiation from an rf excited

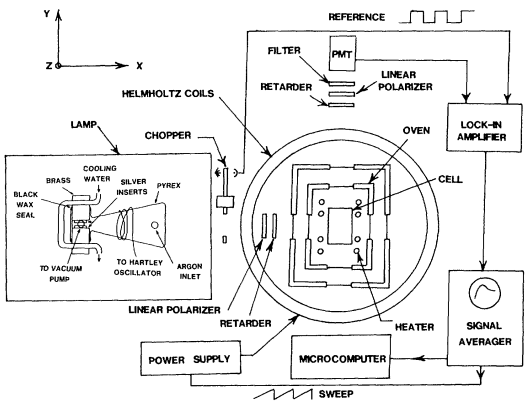


FIG. 2. Details of the experimental arrangement.

silver lamp is chopped at 85 Hz with a mechanical chopper, circularly polarized using a Polaroid HNPB polarizer and a mica quarter-wave retarder, and is focused on the absorption cell containing silver-107 by quartz lenses. The cell is placed in an oven capable of heating the cell to about 1000 K. The 3383-Å fluorescent radiation, spectrally filtered by a 40-Å bandwidth interference filter, and analyzed for circular polarization, is observed by an EMI 9658B photomultiplier tube. In addition, a broadband interference filter is used to further suppress the blackbody radiation from the oven. A phase-sensitive detection using a Princeton Applied Research (PAR) HR-8 lock-in amplifier is done with the reference signal supplied by the chopper. The output of the lock-in amplifier is fed to a Tracor-Northern NS-570 signal averager. A ramp voltage derived from the time base of the signal averager is used to sweep the magnetic field at the cell using a pair of Helmholtz coils. Two additional Helmholtz coil pairs are used to compensate the earth's magnetic field. The averaged signal is viewed on the oscilloscope screen of the signal averager and transferred to a Commodore PET (Personal Electronic Transactor) microcomputer for fitting and storing on a magnetic tape. The 3383-Å radiation transmitted through the cell is also monitored using a 1P28 photomultiplier tube, and this information is used in estimating the temperature of the silver vapor.

Considerable effort was spent in designing a good silver resonance lamp. Several designs were tried including a hollow cathode design. A successful design, which was finally used in our experiment, is similar to that described by Moe,[18] and it is shown in Fig. 2. Two brass plugs are sealed to a pyrex cell by black wax as shown. Silver inserts are soldered on the brass plugs. Ar flows through the cell to keep the silver vapor from depositing on the front window. A couple of turns of wire are wrapped on the cell and connected to a $\sim$70-W Hartley oscillator.[19] An rf discharge is produced in Ar which is quite intense in the region between the two silver electrodes. Silver sputters out of the electrodes due to ion bombardment and silver resonance lines are emitted. The electrodes get very hot and have to be water cooled. In addition air has to be blown on the cell to keep it from getting too hot. We have determined that this lamp is not self-reversed to any significant degree in the following way: We have observed the transmission of the 3383-Å resonance light through

the silver absorption cell. As the cell was heated, we found that the resonance light was absorbed according to Beer's law and about 70% absorption was observed at the highest temperature of the absorption cell that could be conveniently obtained. This upper limit was imposed by the highest temperature that could be attained in our oven, and we saw no evidence of the flattening of absorption curve which would have been indicative of the self-reversal. This behavior was observed even when the lamp was run with maximum rf power from our Hartley oscillator. Our normal operating conditions correspond to rf power much less than the maximum, because the lamp was more stable at lower rf powers. Moreover, it was difficult to keep the silver electrodes from melting at high rf powers.

One of the most difficult technical problems in this experiment was the design of an oven which could heat the silver absorption cell to over 1000 K and meet the stringent requirements of a spectroscopy experiment. For example, it should be made of nonmagnetic materials, the heater current should produce negligible magnetic fields, it should have appropriate windows, thermal gradients in the cell should be such that silver does not condense near the windows, heater black-body radiation should be minimum, etc. After considerable experimentation, we made our oven out of graphite. The heating wire has a coaxial geometry, therefore produces no magnetic field. The oven is placed inside a water-cooled brass vessel which is evacuated to provide good insulation. Details of the oven may be found elsewhere.[20]

The absorption cell itself, about 1-in. diam by 2-in. long, was made of quartz with hard-sealed windows. The cell was evacuated and baked under a vacuum at 1325 K for at least 24 h, sparked by a Tesla coil, and rebaked for another 24 h. Separated isotope of $^{107}$Ag was distilled in the cell at a residual pressure of about $10^{-7}$ Torr. Helium (Matheson research purity grade) was introduced in the cell and the cell was sealed. Helium pressure was read by a Matheson dial gauge just before sealing the cell.

## IV. RESULTS

Figure 3 shows some of our typical data. Figure 3(a) shows the Hanle-effect data with 125 Torr of helium (pressure measured at the seal off temperature of 375 K, see below) while Fig. 3(b) shows the corresponding data without any buffer gas for comparison. Crosses are the experimental data points and the solid lines are the theoretical fits to these data. The theoretical curves were computed using Eq. (3) and were least-squares fitted to the experimental data for several values of $\gamma$. Since we do not measure absolute intensities, the two parameters used in these fits were the vertical scale factor and the zero offset. The residues were plotted against $\gamma$, and the value of $\gamma$ corresponding to the smallest value of the residue was taken to be the best fitted value of $\gamma$. Similar data were taken for several different buffer-gas densities.

In Fig. 4 we have plotted $\gamma$ as a function of the buffer-gas density. The error bars on $\gamma$ represent one standard deviation in statistical spread of the data, contribution of uncertainty in the radiative lifetime of the $5^2P_{1/2}$ state,

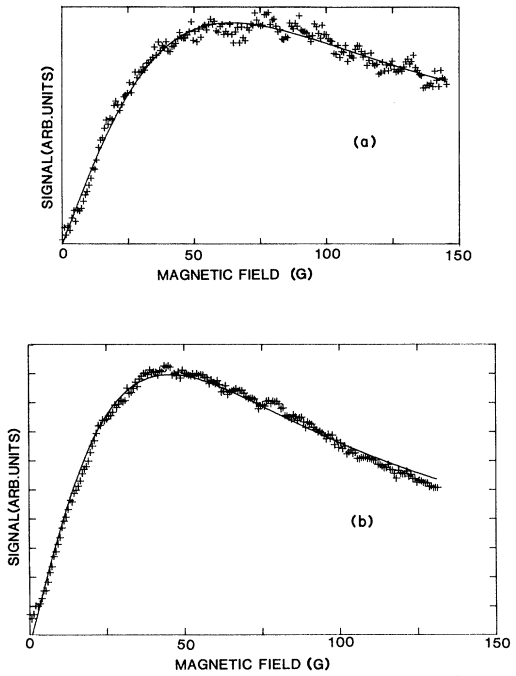


FIG. 3. Hanle-effect signal in the $5^2P_{1/2}$ state of $^{107}$Ag, (a) in the presence of $3.2\times10^{18}$ atoms per cm$^3$ helium, and (b) without any buffer gas. Crosses are the experimental data points and solid lines are the least-squares-fitted theoretical curves computed from Eq. (3).

and uncertainty in the calibration of the magnetic field. Standard deviations are based on four independent data sets for each buffer-gas pressure. The radiative lifetime was measured by the Hanle effect using a cell without buffer gas to be $\tau=7.56(22)$ nsec.[21] This lifetime measurement is in good agreement with the value $\tau=7.5(4)$

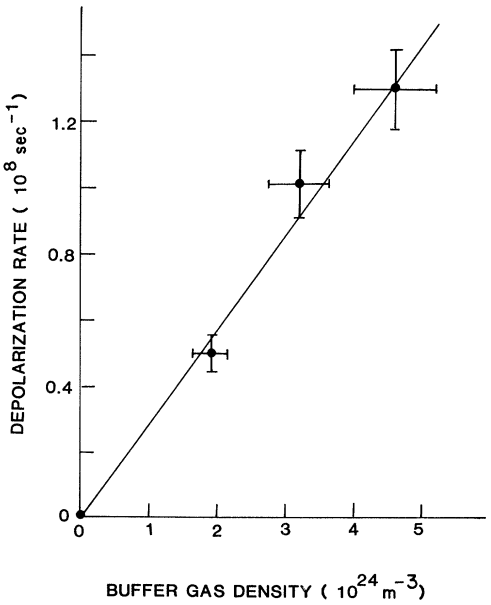


FIG. 4. Electronic depolarization rate $\gamma$ is plotted against the density of helium gas. Depolarization cross section $\sigma$ is obtained from the slope of this curve.

nsec obtained by Cunningham and Link[22] using phase-shift method. The error bar in $\tau$ was taken into account in determining the error bars on $\gamma$. The magnetic field was calibrated using a F.W. Bell, Inc. Model 640 Hall-effect Gaussmeter. The Gaussmeter has a stated calibration accuracy of 1%, therefore we have allowed for 2% uncertainty due to magnetic field measurements. No other source of systematic error could be identified. The data were taken at sufficiently low vapor density of silver ($<10^{11}$ cm$^{-3}$) that no significant coherence narrowing should be present. The vapor density was determined from the measured absorption of the 3383-Å resonance light by silver vapor at the operating temperature. All of our data, including the zero-buffer-gas data, were taken at the same temperature. An evidence that our data does not have significant coherence narrowing is that our result at zero-buffer-gas pressure (radiative lifetime measurement) is in excellent agreement with that of Cunningham and Link[22] obtained by phase-shift method.

The error bars on the buffer-gas density are primarily due to uncertainty in our knowledge of the temperature at which the cells were sealed off. During the process of sealing off, the cell temperature was raised and the *average* temperature of the cell at seal off was estimated from the observed increase in the pressure on the dial gauge and the volume ratio of the gas handling system and the cell. However, there was a large uncertainty in this measurement and we have taken the average temperature of the cell at seal off to be 375±50 K.

The value of the cross section $\sigma$ was deduced using Eq. (6) and the measured slope, $\gamma/n$, from Fig. 4. The error bar in, $\gamma/n$ was determined by visual observation of the plot. In the calculation of $\sigma$, the average relative velocity of the collision partners (Ag-He) at 1025 K was used. The temperature of the vapor was determined from the measured vapor density of silver at the operating temperature (see above) and the published vapor-pressure data.[23] Since the vapor-pressure data for silver have large uncertainties, an error bar of ±75 K was assigned to the temperature measurement. Corresponding uncertainty in the average relative velocity is about ±4%. Using the above procedure, we obtain for the nuclear-spin-independent depolarization cross section of the silver $5^2P_{1/2}$ state by helium

$$\sigma = 1.2 \pm 0.15 \times 10^{-16} \text{ cm}^2 .$$

TABLE I. Comparison of the fine-structure intervals and the cross sections for depolarization of $P_{1/2}$ states by He for several single-valence-electron atoms.

| Element (state) | Fine-structure interval $\Delta E$ (cm$^{-1}$) | Depolarization cross section (cm$^2$) | $\sigma(\Delta E)^2$ |
|---|---|---|---|
| Rb($5^2P_{1/2}$) | 237 | $24\times10^{-16}$[a] | $1.35\times10^{-10}$ |
| Cs($6^2P_{1/2}$) | 554 | $6.1\times10^{-16}$[b] | $1.87\times10^{-10}$ |
| Ag($5^2P_{1/2}$) | 921 | $1.2\times10^{-16}$[c] | $1.02\times10^{-10}$ |
| Tl($6^2P_{1/2}$) | 7793 | $6\times10^{-19}$[d] | $0.36\times10^{-10}$ |

[a]Reference 3.
[b]Reference 7, corrected according to Ref. 3.
[c]This work.
[d]Reference 24.

The above cross section is more than an order of magnitude smaller than the corresponding cross section in the $5^2P_{1/2}$ state of Rb ($\sigma=24\times10^{-16}$ cm$^2$).[3] Anomalously small cross section for silver is perhaps understandable if the depolarization takes place predominantly due to electrostatic interaction.[1,3,7,13,14,24] In that case the depolarization proceeds via virtual transition to the $5^2P_{3/2}$ state. Therefore the $P_{1/2}$ state depolarization cross section should be inversely proportional to the square of fine-structure interval $\Delta E$. Since the fine-structure interval in Ag is 921 cm$^{-1}$ while it is only 237 cm$^{-1}$ in Rb, we expect the cross section in Ag to be smaller by a factor of 15 due to this reason alone. In Table I we have listed the depolarization cross sections for $P_{1/2}$ states of several single-valence-electron atoms. Included in this table is the depolarization cross section of Tl,[24] which has a very large fine-structure interval in the ground $6^2P$ state. The last column of the table lists $\sigma(\Delta E)^2$ and they are found to be within an order of magnitude of each other, in accord with our expectations.

## ACKNOWLEDGMENTS

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