

# Orthopositronium annihilation rates in gaseous halogenated methanes

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The normalized orthopositronium quenching rate,  ${}_1Z_{\text{eff}}$ , has been measured for  $\text{CH}_3\text{F}$ ,  $\text{CH}_3\text{Cl}$ ,  $\text{CH}_3\text{Br}$ , and  $\text{CH}_3\text{I}$  in the gas phase. Silica aerogel was used as the positronium formation medium and the microchambers. The  ${}_1Z_{\text{eff}}$  values obtained are  ${}_1Z_{\text{eff}}(\text{CH}_3\text{F}) = 0.46 \pm 0.03$ ,  ${}_1Z_{\text{eff}}(\text{CH}_3\text{Cl}) = 0.58 \pm 0.05$ ,  ${}_1Z_{\text{eff}}(\text{CH}_3\text{Br}) = 0.70 \pm 0.04$ , and  ${}_1Z_{\text{eff}}(\text{CH}_3\text{I}) = 2.1 \pm 0.2$ . The  ${}_1Z_{\text{eff}}$  value for  $\text{CH}_3\text{I}$  is significantly larger than expected for pick-off quenching. It is probably caused by spin-conversion quenching through spin-orbit interaction.

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## I. INTRODUCTION

Positronium (Ps) is the hydrogenlike state consisting of a positron-electron pair. The spin-triplet positronium, orthopositronium (*o*-Ps), which self-annihilates into  $3\gamma$  with the annihilation rate of  $\lambda_{3\gamma}$  ( $=1/142$  ns) in vacuum, can be quenched into  $2\gamma$  through collisions with atoms and molecules. In a gas of density  $n$ , the annihilation rate of *o*-Ps is written as

$$\lambda_{o\text{-Ps}}^{\text{total}}(n) = \lambda_{3\gamma} + \lambda_{\text{gas}}(n), \quad (1)$$

where  $\lambda_{\text{gas}}(n)$  is the rate of collisional quenching on the molecules. In a moderate pressure range,  $\lambda_{\text{gas}}(n)$  depends linearly on  $n$  for most molecules. To quantify the quenching abilities of molecules, we introduce a dimensionless parameter

$${}_1Z_{\text{eff}} \equiv \frac{\lambda_{\text{gas}}}{4\pi r_0^2 cn}, \quad (2)$$

where  $4\pi r_0^2 c$  ( $r_0$  is the classical electron radius and  $c$  the speed of light) is the Dirac  $2\gamma$  annihilation rate for a spin-singlet positron-electron pair [1,2]. This parameter was originally introduced as “ ${}_1Z_{\text{eff}}$ ,” with the superscript “1,” for the pick-off quenching process in which the positron of the Ps annihilates into  $2\gamma$  with a foreign electron that pairs in a spin-singlet state with the positron [3].

Four categories of quenching are known so far: (i) pick-off quenching [1,3], (ii) spin-conversion quenching through electron exchange [4–7], (iii) attachment (chemical) quenching [8–10], and (iv) spin-conversion quenching through spin-orbit interaction, which was found recently [11,12]. Most of the gas species quench Ps only through process (i) with  ${}_1Z_{\text{eff}}(\text{pick-off}) \sim 0.1\text{--}1$ . Only a few species have been found to quench Ps through processes (ii) with  ${}_1Z_{\text{eff}}(\text{electronexchange}) \sim 40\text{--}200$ , (iii) with

${}_1Z_{\text{eff}}(\text{attach}) \sim 10^4\text{--}10^6$ , and (iv) with  ${}_1Z_{\text{eff}}(\text{spin-orbit}) \sim 0.1\text{--}0.8$ , in addition to the pick-off quenching.

Positronium-molecule interactions in the gas phase have been investigated widely using the sample gas as the Ps formation medium (the gas method) [1,13–15]. With this method, however, measurements of  ${}_1Z_{\text{eff}}$  at room temperature are difficult to make for gases with low vapor pressure. At low pressures, the stopping power of the gas is so small that the positrons from the source reach the inner wall of the experimental chamber, where they annihilate, and hence the Ps formation fraction is much reduced. Moreover, the free positrons that do not form Ps may survive for a longer time, and the annihilation signals from these may affect the single-exponential analysis of the *o*-Ps decay rate [14]. Thus, measurements of the *o*-Ps annihilation rate in low-vapor-pressure gases at room temperature have been performed on only a few species [16–18] having high stopping powers and large annihilation cross sections for the free positron, and for the purpose of determining the self-annihilation rate of *o*-Ps in vacuum.

A different method of investigating Ps-molecule interactions using silica aerogel as the Ps formation medium and microchambers has been developed [5,7,12,19–25]. In this method, about half of the positrons injected into the silica aerogel, a three-dimensional network of silica nanograins, form Ps [22]. Furthermore, positrons that do not form Ps stay inside the grains after thermalization because of the positive work function and annihilate quickly so that they do not affect the *o*-Ps lifetime spectrum. Because of these properties, silica aerogel has been used in obtaining the values of  ${}_1Z_{\text{eff}}$  for various atoms and molecules in the gas phase [7,12].

Recently, we have developed a measurement and analysis method for obtaining the  ${}_1Z_{\text{eff}}$  value for low-vapor-pressure gases [26]. It incorporates an extrapolation technique addressing the effect of molecular adsorption on the surface of the silica aerogel.

In the present work, we have employed this technique of measurement and this analysis method to obtain the values of  ${}_1Z_{\text{eff}}$  for halogenated methanes in the gas phase.

## II. EXPERIMENT

The experimental arrangement and analysis method employed in this work for determining the thermally averaged

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TABLE I. The purities and the suppliers of samples investigated.

Sample	Purity	Supplier
CH <sub>4</sub>	99.999%	Toyoko Kagaku Co., Ltd
CH <sub>3</sub> F	99.9%	Toyoko Kagaku Co., Ltd
CH <sub>3</sub> Cl	99.99%	Shinetsu Chemical Co., Ltd.
CH <sub>3</sub> Br	99.5%	Sanko Chemical Industry Co., Ltd.
CH <sub>3</sub> I	99.5%	Sigma-Aldrich Industry Co.

values of  ${}_1Z_{\text{eff}}$  is described in detail elsewhere [26]. Briefly, a 30 kBq  $^{22}\text{Na}$  positron source, sealed with two strips of  $7.5\ \mu\text{m}$  Kapton polyimide film, was sandwiched between two pieces of silica aerogel of size  $10 \times 10 \times 5\ \text{mm}^3$ . Hydrophobic silica aerogels were used, namely, SP-15, YI-30, and SP-50, supplied by Panasonic Corporation. These silica aerogels are produced by  $-\text{Si}(\text{Me})_3$  [trimethylsilyl substituent (TMS)] modification of alcogels followed by  $\text{CO}_2$  supercritical drying [27]. The surface properties of these aerogels are identical with each other; only the radii of the  $\text{SiO}_2$  nanograins and the mean free distances between the grains are different. The surface properties are more stable than in other types.

The gases investigated were  $\text{CH}_4$ ,  $\text{CH}_3\text{F}$ ,  $\text{CH}_3\text{Cl}$ ,  $\text{CH}_3\text{Br}$ , and  $\text{CH}_3\text{I}$ . Although the  ${}_1Z_{\text{eff}}$  value of  $\text{CH}_4$  has been measured by the gas method [28,29], we included  $\text{CH}_4$  in our study as a comparison. The vapor pressures of these gases are 3.3 ( $\text{CH}_3\text{F}$ ), 0.6 ( $\text{CH}_3\text{Cl}$ ), 0.19 ( $\text{CH}_3\text{Br}$ ), and 0.053 MPa ( $\text{CH}_3\text{I}$ ) at room temperature. Table I lists the purities and the suppliers of these gases. The samples of  $\text{CH}_4$ ,  $\text{CH}_3\text{F}$ , and  $\text{CH}_3\text{Cl}$  were supplied in a standard gas cylinder and, in the experiment, the measurement chamber was simply filled with these gases following evacuation. However, the  $\text{CH}_3\text{Br}$  and  $\text{CH}_3\text{I}$  samples were supplied as liquids in a sealed can and a sealed glass bottle, respectively. The liquid samples were deoxygenated by the standard freeze-pump-thaw method and vaporized into the measurement chamber.

The decay rates of the positrons were measured over a range of gas densities using each of the silica aerogels and the gases in order of increasing gas density. The chamber was evacuated between measurements at one density and the next. Room temperature was recorded during the measurements at  $300 \pm 1\ \text{K}$  for  $\text{CH}_3\text{Cl}$ ,  $296 \pm 1\ \text{K}$  for  $\text{CH}_3\text{I}$ , and  $298 \pm 1.5\ \text{K}$  for the other species.

The number density of the gas molecules was calculated from the gas pressure measured by a capacitance manometer and temperature using the second virial coefficient.

### III. RESULTS AND DISCUSSION

Figure 1 shows the positron lifetime spectrum for  $\text{CH}_3\text{Br}$  at 0.04 MPa with aerogel YI-30. A sharp prompt peak is followed by a slow-decay component and a flat background. All the signals from non-Ps positrons as well as those from the spin-singlet parapositronium ( $p$ -Ps), are included in the prompt peak. The flat background is due to chance coincidences.

The total annihilation rate of  $o$ -Ps in gases in this system can be written as

$$\lambda_{o\text{-Ps}}^{\text{total}} = \lambda_{3\gamma} + \lambda_{\text{gas}} + \lambda_{\text{SiO}_2}, \quad (3)$$

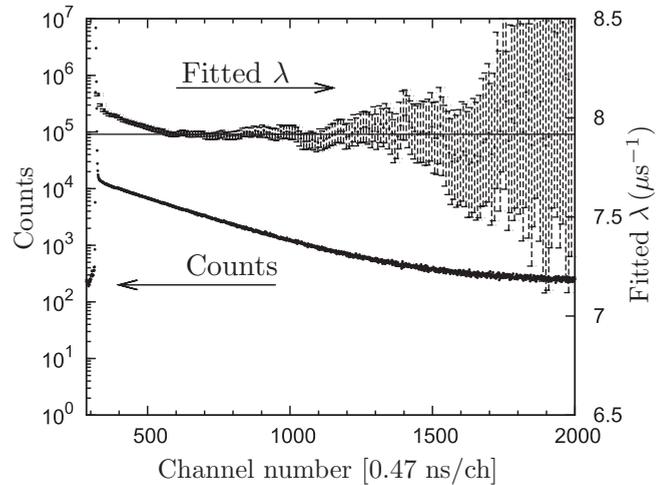


FIG. 1. Positron lifetime spectrum for  $\text{CH}_3\text{Br}$  at 0.04 MPa in silica aerogel YI-30. The upper set of points with uncertainty bars show the  $\lambda$  extracted from the fit of  $(Ae^{-\lambda t} + B)e^{-Rt}$  to the distribution, plotted as a function of the starting channel for the fit.

where  $\lambda_{\text{SiO}_2}$  is the quenching rate on the silica grain surfaces. The measured spectrum for a single-exponential decay with a constant decay rate  $\lambda$  has the form  $(Ae^{-\lambda t} + B)e^{-Rt}$ , where  $A$  and  $B$  are constants,  $t$  is time, and  $R$  is the stop rate that is measured independently [30,31]. We fitted this function to the obtained spectra, progressively stepping out the starting channel of the fit to determine the decay rate of thermalized  $o$ -Ps [32]. The variation of the extracted value of  $\lambda$  is shown in Fig. 1 as a function of the starting channel of the fit. For all the spectra obtained, the values of the extracted  $\lambda$  with the starting channel corresponding to  $t \geq 170\ \text{ns}$  were constant within the statistical uncertainty. The values of the fitted  $\lambda$  at  $t = 170\ \text{ns}$  were thus taken to be the total annihilation rate,  $\lambda_{o\text{-Ps}}^{\text{total}}$ , of the thermalized  $o$ -Ps.

Figure 2 shows the  $\lambda_{o\text{-Ps}}^{\text{total}}$  values obtained with the silica aerogels SP-15, YI-30, and SP-50 plotted against the number density  $n$  of the gases. Because of changes in legal requirements relating to the use of  $\text{CH}_3\text{Cl}$ , it was only possible to obtain the set of data shown in Fig. 2 for this gas. The vapor pressure of  $\text{CH}_3\text{I}$  at room temperature is 0.053 MPa and so measurements above 0.040 MPa (0.37 amagat) were not made with samples YI-30 and SP-50. Additionally, the maximum pressure plotted with aerogel SP-15 in Fig. 2 is 0.027 MPa (0.24 amagat); a drastic destructive shrinkage in macroscopic volume of the SP-15 sample of about 40% was noticed after the measurements. Abnormally large and nonreproducible annihilation rates were obtained at 0.033 MPa (0.30 amagat) and 0.040 MPa (0.37 amagat), which are not included in Fig. 2.

The gradients of the sets of data for  $\text{CH}_3\text{Br}$  and  $\text{CH}_3\text{I}$  in Fig. 2 vary slightly, in contrast to the data for  $\text{CH}_4$  and  $\text{CH}_3\text{F}$ . This indicates that  $\lambda_{\text{SiO}_2}$  also depends on gas density and is attributed to the adsorption of the gas molecules on the grain surfaces [26].

Since the surface properties of the TMS-modified aerogels used are identical, modification of the surfaces by gas absorption is the same. Therefore, the Ps quenching probability per collision on the grain surfaces is the same at the same gas

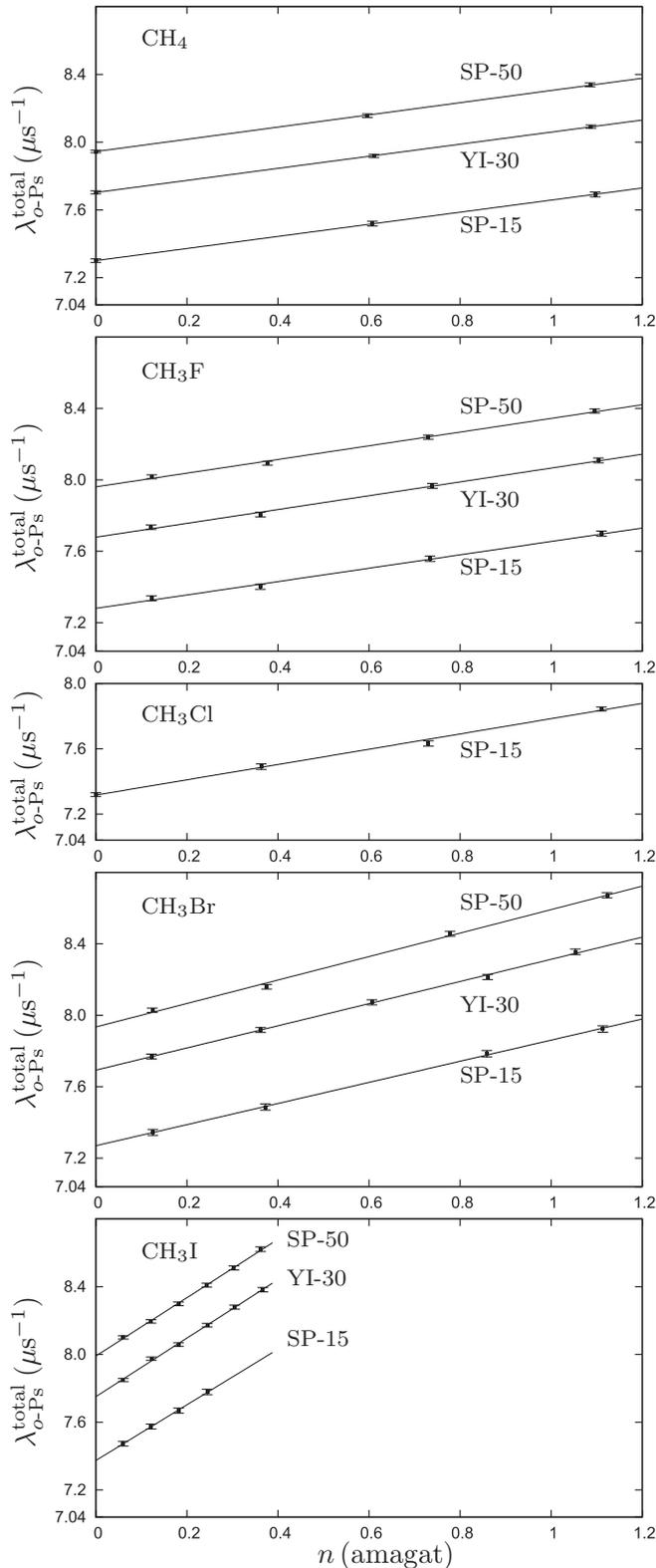


FIG. 2. Values of  $\lambda_{o\text{-Ps}}^{\text{total}}$  plotted as a function of the number density  $n$  for  $\text{CH}_4$ ,  $\text{CH}_3\text{F}$ ,  $\text{CH}_3\text{Cl}$ ,  $\text{CH}_3\text{Br}$ , and  $\text{CH}_3\text{I}$  ( $1 \text{ amagat} \simeq 2.69 \times 10^{25} \text{ molecule m}^{-3}$ , the density of an ideal gas at 273 K and 1 atm).

density. Thus  $\lambda_{\text{SiO}_2}$ , which is the product of the Ps collision rate and the quenching probability per collision on the grain surface, is expected to be proportional to the rate of Ps collision

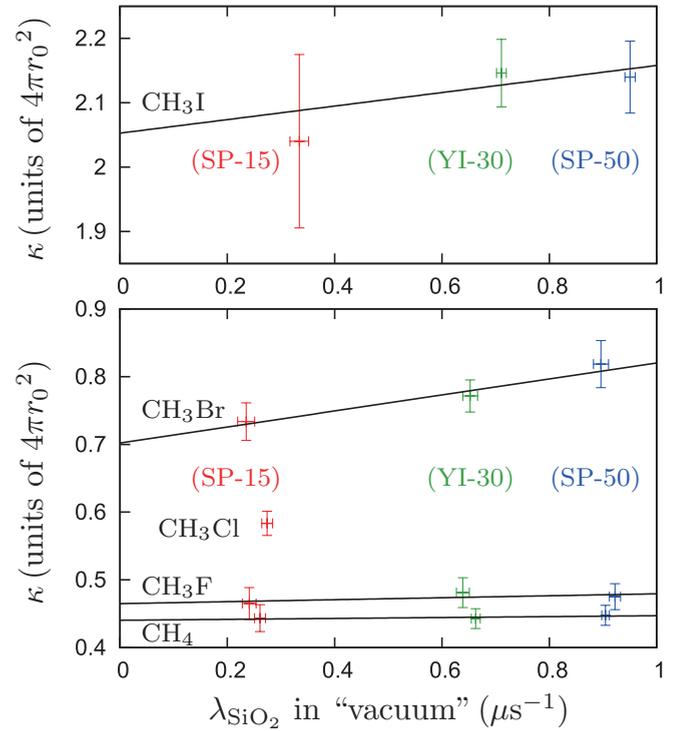


FIG. 3. (Color online) Gradients  $\kappa$  of the fitted straight lines in Fig. 2 plotted against  $\lambda_{\text{SiO}_2}$  values in “vacuum” for the aerogels used. The  $\lambda_{\text{SiO}_2} \rightarrow 0$  limits correspond to the true  ${}_1Z_{\text{eff}}$ .

on the grain surfaces at the same gas density. We used  $\lambda_{\text{SiO}_2}$  in “vacuum,” which is the  $n \rightarrow 0$  intercept for each aerogel in Fig. 2, as a measure of the collision rate of Ps on the grain surfaces of the TMS-modified aerogels [26].

Figure 3 shows the gradient  $\kappa$  of each of the fitted straight lines in Fig. 2 as a function of the  $\lambda_{\text{SiO}_2}$  value in “vacuum” for the aerogels used. A straight line was fitted to the data as shown in Fig. 3. The extrapolation limit  $\lambda_{\text{SiO}_2} \rightarrow 0$  for each fitted line corresponds to a zero collision rate on the grain surfaces. We have derived the  ${}_1Z_{\text{eff}}$  values from this extrapolation as  ${}_1Z_{\text{eff}}(\text{CH}_4) = 0.44 \pm 0.03$ ,  ${}_1Z_{\text{eff}}(\text{CH}_3\text{F}) = 0.46 \pm 0.03$ ,  ${}_1Z_{\text{eff}}(\text{CH}_3\text{Br}) = 0.70 \pm 0.04$ , and  ${}_1Z_{\text{eff}}(\text{CH}_3\text{I}) = 2.1 \pm 0.2$ . The result for  $\text{CH}_4$  is consistent with those from previous work with the gas method [28,29].

For  $\text{CH}_3\text{Cl}$ , we obtained the data only with the SP-15 aerogel and could not employ the extrapolation method described above. From the value of  $\kappa$  for  $\text{CH}_3\text{Cl}$  obtained with SP-15, we determined  ${}_1Z_{\text{eff}}(\text{CH}_3\text{Cl}) = 0.58 \pm 0.05$  with an arbitrarily assigned uncertainty.

Plotted in Fig. 4 are  ${}_1Z_{\text{eff}}$  values against the geometric cross section estimated using  $\pi(d/2)^2 = M\bar{v}/8\sqrt{2}\eta$ , where  $d$  is the diameter of the molecule,  $M$  is the mass of the molecule,  $\bar{v}$  is the averaged absolute velocity of the molecule, and  $\eta$  is the viscosity in the gas phase, which depends on the temperature of the gas [33]. Reported values for noble gases [1,34,35] are also plotted in Fig. 4. Recently, it was revealed that Kr and Xe quench  $o\text{-Ps}$  via spin conversion caused by spin-orbit interaction [12]. The annihilation rate was decomposed into the pick-off quenching rate  $\lambda_{\text{pick-off}}$  and the spin-conversion quenching rate via spin-orbit interaction,  $\lambda_{\text{spin-orbit}}$ . In Fig. 4, the values  ${}_1Z_{\text{eff}}(\text{pick-off}) = \lambda_{\text{pick-off}}/4\pi r_0^2 cn$  for Kr and

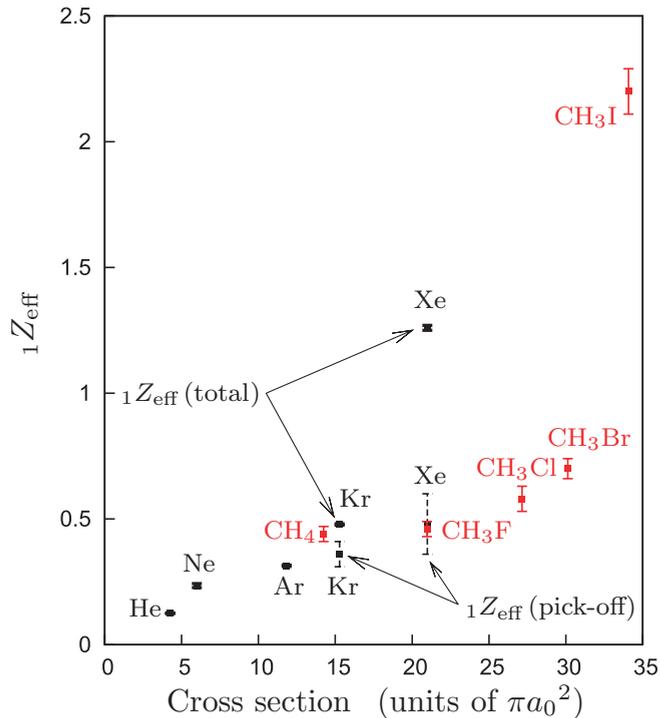


FIG. 4. (Color online)  ${}_1Z_{\text{eff}}$  values obtained for methane and halogenated methanes plotted against the geometric cross sections of the target molecules, which were estimated from viscosity. Previous values of  ${}_1Z_{\text{eff}}$  for He, Ne [34], Ar [1], and  ${}_1Z_{\text{eff}}(\text{total})$  and  ${}_1Z_{\text{eff}}(\text{pick-off})$  for Kr and Xe [1,12,35], are also plotted.

Xe are plotted with dashed uncertainty lines, as well as  ${}_1Z_{\text{eff}}(\text{total}) = \lambda_{\text{gas}}/4\pi r_0^2 cn$  for these atoms. The values of  ${}_1Z_{\text{eff}}(\text{pick-off})$  for Kr and Xe are roughly proportional to the geometric cross section along with the  ${}_1Z_{\text{eff}}$  values of the other atoms or molecules except CH<sub>3</sub>I.

The  ${}_1Z_{\text{eff}}$  value obtained for CH<sub>3</sub>I is extraordinarily large for pick-off quenching and indicates the presence of another mechanism in the process. It cannot be accounted for by spin-conversion quenching via electron exchange since this reaction is possible only when either the initial or final electronic state of the target is nonsinglet. This is not the case in Ps-CH<sub>3</sub>I collisions at thermal energies ( $\sim 0.04$  eV), because the ground state of CH<sub>3</sub>I is singlet and the lowest excitation energy of CH<sub>3</sub>I is larger than 6 eV. Attachment quenching is not likely either, considering the  ${}_1Z_{\text{eff}}$  value obtained. In this quenching process, the positron of the Ps stays in the vicinity of the target molecule for a considerable period, resulting in an extremely high annihilation rate. Known attachment quenching yields,  ${}_1Z_{\text{eff}}(\text{attachment})$ , are  $\sim 10^4$ – $10^6$ , which is several orders of

magnitude higher than the value obtained of  ${}_1Z_{\text{eff}}(\text{CH}_3\text{I}) = 2.1$ .

Spin-conversion quenching via spin-orbit interaction is the most likely explanation. The spin-conversion cross section  $\sigma_{\text{so}}(k)$  for the  $o$ -Ps  $\rightarrow$   $p$ -Ps reaction via spin-orbit interaction is parametrized as  $\sigma_{\text{so}}(k) = F_{\text{so}}\sigma_p(k)$ , where  $F_{\text{so}}$  is the conversion probability per elastic  $p$ -wave collision, and  $\sigma_p(k)$  is the elastic  $p$ -wave cross section [11] [the value of  ${}_1Z_{\text{eff}}(\text{spin-orbit})$  at thermal velocity is proportional to  $\sigma_{\text{so}}$ ]. The  $F_{\text{so}}$  value is expected to scale roughly as the fourth power of the atomic number [11]. Since the atomic number of I is only one smaller than that of Xe, it is conceivable that the spin-conversion process via spin-orbit interaction takes place in the Ps-CH<sub>3</sub>I collision.

The  ${}_1Z_{\text{eff}}(\text{total})$  values for Xe and CH<sub>3</sub>I appear to be proportional to the geometric cross section. If the  $F_{\text{so}}$  value for CH<sub>3</sub>I is almost the same as that of Xe and the cross section for the elastic  $p$ -wave collision is roughly proportional to the geometric cross section estimated from viscosity, the  ${}_1Z_{\text{eff}}(\text{spin-orbit})$  value will also be proportional to the geometric cross section. This suggests that the  ${}_1Z_{\text{eff}}(\text{pick-off})$  value for CH<sub>3</sub>I is not much different from the  ${}_1Z_{\text{eff}}(\text{total})$  value for CH<sub>3</sub>Br.

The decomposition of the value of  ${}_1Z_{\text{eff}}(\text{total})$  into  ${}_1Z_{\text{eff}}(\text{pick-off})$  and  ${}_1Z_{\text{eff}}(\text{spin-orbit})$  components will be possible by positron lifetime measurement in a magnetic field, as reported previously [12], or by the age-momentum correlation (AMOC) measurement of positron annihilation [7]. We are now preparing to make these measurements to explain the extraordinarily large  ${}_1Z_{\text{eff}}$  value of CH<sub>3</sub>I.

#### IV. CONCLUSION

The thermally averaged values of  ${}_1Z_{\text{eff}}$  were experimentally determined for CH<sub>3</sub>F, CH<sub>3</sub>Cl, CH<sub>3</sub>Br, and CH<sub>3</sub>I in the gas phase using silica aerogels as highly efficient Ps formation media and microchambers. The values obtained were  ${}_1Z_{\text{eff}}(\text{CH}_3\text{F}) = 0.46 \pm 0.03$ ,  ${}_1Z_{\text{eff}}(\text{CH}_3\text{Cl}) = 0.58 \pm 0.05$ ,  ${}_1Z_{\text{eff}}(\text{CH}_3\text{Br}) = 0.70 \pm 0.04$ , and  ${}_1Z_{\text{eff}}(\text{CH}_3\text{I}) = 2.1 \pm 0.2$ . The value of  ${}_1Z_{\text{eff}}(\text{CH}_3\text{I})$  was found to be significantly larger than expected for pick-off quenching. It is suggested that this is because of spin-conversion quenching through spin-orbit interaction.

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