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Sensitivity of Orthopositronium Annihilation Rates to Density Fluctuations in Ethane Gas

Suresh C. Sharma, Abbas Eftekhari, and John D. McNutt[(a)]

Center for Positron Studies, The University of Texas at Arlington, Arlington, Texas 76019

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Orthopositronium annihilation rates have been measured in ethane gas for densities in the range 1.8 to 286 amagats at 306.4 K. The behavior of the measured annihilation rates at densities up to about 70 amagats can be explained in terms of a simple density fluctuation model. New features are observed at higher densities of ethane at 306.4 K.

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In this Letter we present results from a continuing series of experiments whereby positron annihilation techniques are used to investigate the influence of density fluctuations on the annihilation of orthopositronium (o-Ps) atoms formed in ethane gas. We present new results that are the first to show a complex dependence of orthopositronium annihilation rates ($\lambda_{o\text{-Ps}}$) on the density of the gas over a wide range of densities at a temperature that is only about 1 K above the critical temperature of the gas. These results support the sensitivity of $\lambda_{o\text{-Ps}}$ to density fluctuations in ethane gas and allow us to study the application of a simple density fluctuation model to $\lambda_{o\text{-Ps}}$ at sufficiently high densities of the gas that density fluctuations are expected to be large and highly correlated. The behavior of o-Ps annihilation rates has been reported recently as a function of density and temperature in methane[1] and ethane[2] gases. However, these measurements were made only for limited ranges of densities and temperatures of the gases. Nevertheless, they have indicated that the annihilation rates of o-Ps atoms in methane and ethane gases are sensitive to density fluctuations. Generally at low densities, o-Ps annihilation rates increase linearly with the density of the gas up to certain temperature-dependent values of gas density

(D_1*), according to

$$\lambda_{o\text{-Ps}} = \lambda_{\text{vac}} + 4\pi r_0{}^2 c n_0 \, {}^1Z_{\text{eff}} D. \qquad (1)$$

$\lambda_{\text{vac}} = 7.06 \times 10^{-3}$ nsec^{-1} is the annihilation rate of o-Ps atoms in vacuum,[3] r_0 is the classical radius of the electron, c is the speed of light, n_0 is the standard number density, and D is the density of the gas in amagats. ${}^1Z_{\text{eff}}$ is an empirical parameter that is defined as effective number of electrons per molecule available for the pickoff annihilation of o-Ps atoms. The measured values of $\lambda_{o\text{-Ps}}$ at densities higher than D_1*, in methane and ethane gases, are observed to be significantly smaller than those values calculated from Eq. (1) for the same density and temperature of the gas. The deviations ($\Delta\lambda$) of the measured annihilation rates of o-Ps atoms from Eq. (1), for small ranges of densities higher than D_1*, have been correlated with fractional deviations of the density of the gas from ideal-gas density at the same pressure and temperature and with average values of relative density fluctuations in the gas. In addition, the observed behavior of $\lambda_{o\text{-Ps}}$ with temperature in xenon gas near its critical temperature[4] and a temperature-dependent behavior of ${}^1Z_{\text{eff}}$ in ethane gas[2] have also been attributed to density fluctuations in these gases. However, these results are understood only qualitatively. In order to understand better the complex process(es) of the annihilation of o-Ps atoms we have obtained data for $\lambda_{o\text{-Ps}}$ in ethane gas at 306.4 ± 0.5 K ($T_c = 305.33$ K)[5] for a range of densities from about 1.8 to 286 amagats which is approximately four times higher than the maximum density of ethane possible in earlier measurements.

Positron lifetime spectra were measured with a standard fast-slow timing spectrometer with a full width at half maximum of about 0.45 nsec for ^{60}Co prompt resolution data. Commercially supplied research-grade ethane with a minimum purity of 99.96 mole% in the liquid phase[6] was used in a carefully cleaned stainless-steel experimental chamber that was immersed in a temperature-controlled oil bath. The lifetime spectra were analyzed generally into three components by using POSITRONFIT EXTENDED.[7] The longest-lived component with an annihilation rate $\lambda_{o\text{-Ps}}$ is assumed to result from pickoff annihilations of o-Ps atoms. The details of the measurements of temperature and pressure of the gas, the lifetime spectra, calculations of the density of the gas, and data analyses have been discussed elsewhere.[8,9]

$\lambda_{o\text{-Ps}}$ measured in ethane gas at 306.4 ± 0.5 K is plotted as a function of density of the gas in Fig. 1. At densities lower than about 11 amagats, $\lambda_{o\text{-Ps}}$ follows Eq. (1) with a ${}^1Z_{\text{eff}} = 0.625 \pm 0.020$. ${}^1Z_{\text{eff}}$ has recently been observed to decrease with increasing temperature of ethane gas between 256 and 377 K.[2] Following this observation, a linear dependence of ${}^1Z_{\text{eff}}$ with temperature has been assumed. The values of $\lambda_{o\text{-Ps}}$ measured for ethane densities higher than about 11 amagat deviate significantly from those values calculated from Eq. (1) for the same density and temperature of the gas. For the sake of simplicity, the observed density dependence of the o-Ps annihilation rates can be divided, somewhat arbitrarily, into three regions: a low-density region where $\lambda_{o\text{-Ps}}$ follows Eq. (1) up to about 11 amagats, an intermediate region from about 11 to 180 amagats where $\lambda_{o\text{-Ps}}$ continues to deviate farther from Eq. (1) and seems to reach a plateau at around 120 amagats, and a high-density region above approximately 180 amagats where $\lambda_{o\text{-Ps}}$ increases almost linearly with ethane density. The measurements of $\lambda_{o\text{-Ps}}$ for ethane densities higher than 286 amagats were not possible because of a limited range of pressure gauges available for the present measurements.

Following the simple density fluctuation model developed recently to explain the observed density and temperature dependencies of $\lambda_{o\text{-Ps}}$ in methane,[1] we define $\Delta\lambda$ as deviations of the meas-

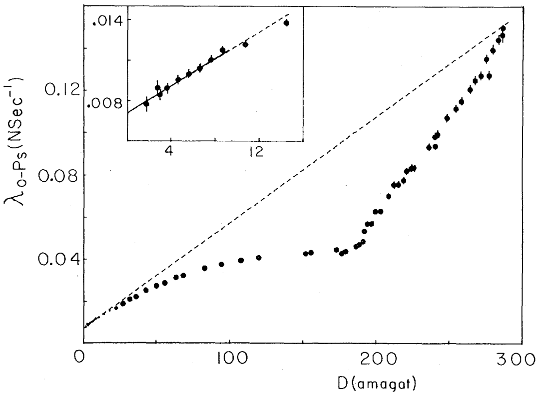

FIG. 1. Orthopositronium annihilation rates vs ethane gas density at 306.4 K. The statistical standard deviations of the annihilation rates fall within the size of the data points unless otherwise shown. For reasons of clarity, the data points at low densities have been reduced in size. The solid line represents a weighted least-squares fit to Eq. (1) up to 10.7 amagats and constrained to pass through λ_{vac} at zero density. The dashed line is an extrapolation of this fit. The inset shows the low-density data.

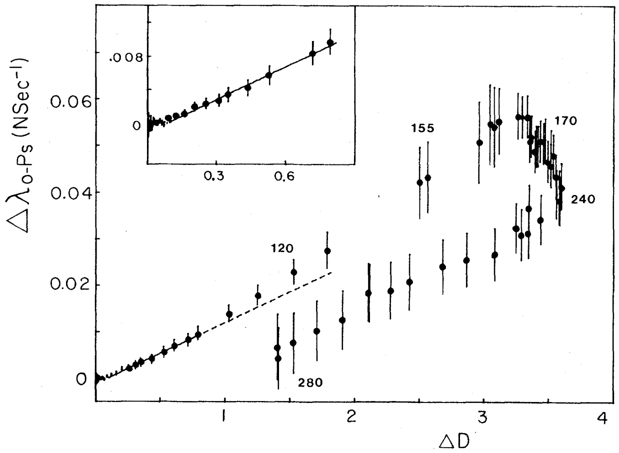

FIG. 2. Deviations $\Delta\lambda$ of the measured orthopositronium annihilation rates from those rates calculated from Eq. (1) at the same density and temperature, vs the fractional deviations ΔD of the gas density from ideal-gas density at 306.4 K. The statistical standard deviations fall within the size of the data points unless otherwise shown. The line represents a weighted least-squares fit to data for a range of densities corresponding to $0.13 < \Delta D < 0.8$. As an aid to the eye, letters indicate the direction of increasing density. The inset shows data for low densities.

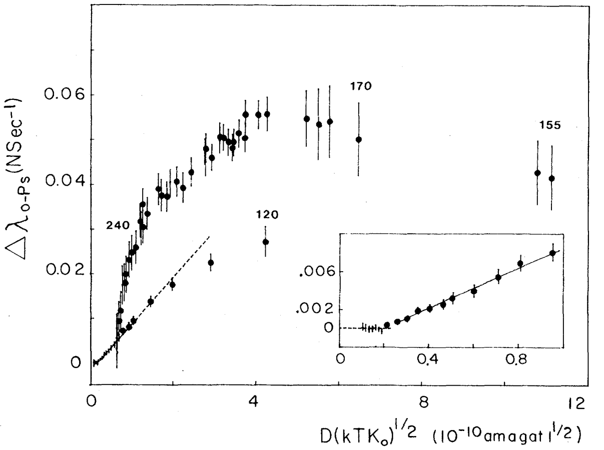

FIG. 3. Deviations $\Delta\lambda$ vs $D(kTK_0)^{1/2}$ at 306.4 K. The statistical standard deviations fall within the size of the points unless otherwise shown. The line represents a weighted least-squares fit to the data for $D(kTK_0)^{1/2}$ values corresponding to $11 \lesssim D \lesssim 68$ amagats. As an aid to the eye, letters indicate the direction of increasing density. The inset shows the low-density data.

ured values of $\lambda_{o\text{-Ps}}$ from Eq. (1) at the same density and temperature. We also define $\Delta D = (D - D_I)/D_I$, where D and D_I are the average density of the gas and ideal-gas density, respectively, at the same pressure and temperature. Using these definitions we calculate $\Delta\lambda$ and ΔD for the data shown in Fig. 1. The resulting data are shown in Fig. 2. $\Delta\lambda = 0$ for a range of densities with a temperature-independent value of the upper limit of this range given by $\Delta D^* \sim 0.1$, which has been observed to be approximately the same for both methane and ethane gases.[1,2] $\Delta\lambda$ appears to increase linearly with ΔD in a temperature-independent manner at densities corresponding to ΔD values from 0.1 to about 0.8 in ethane at 306.4 K and to about 0.5 in methane. The data presented here show that at sufficiently high densities of ethane corresponding to $\Delta D > 0.8$ at 306.4 K, $\Delta\lambda$ varies with ΔD in a manner that is much more complex than that seen previously.

We consider that o-Ps atoms "sample" time-averaged density fluctuations in a small cell of equilibrium volume V_0 that is in thermal contact with the rest of the experimental chamber at a temperature T. The relative density fluctuation δ is defined by $\delta = (D' - D)/D$, where D' and D represent a particular and equilibrium densities, respectively. The probability of a relative density fluctuation between δ and $\delta + d\delta$ in the cell is then given by[10]

$$W(\delta)d\delta = C\exp(-L/kT)d\delta, \tag{2}$$

where C is a normalization constant, k is the Boltzmann constant, and $L = -\int_{V_0}^{V}(P - P_0)dV$ is the work done on the gas in the cell in creating a fluctuation δ. On expansion of the pressure of the gas about its equilibrium value in a Taylor series, the probability distribution of Eq. (2) is given by

$$W(\delta)d\delta = C\exp\left[\frac{V_0^2\delta^2}{2kT}\left(\frac{\partial P}{\partial V}\right)_T - \frac{V_0^3\delta^3}{6kT}\left(\frac{\partial^2 P}{\partial V^2}\right)_T + \frac{V_0^4\delta^4}{24kT}\left(\frac{\partial^3 P}{\partial V^3}\right) + \dots\right]. \tag{3}$$

Under the assumption that o-Ps atoms "sample" regions in which the time-averaged density is lower than the equilibrium density of the gas and that V_0 is independent of gas density, $\Delta\lambda$ values at low densities ($D \gtrsim D_1^*$) can be calculated by keeping only the first term in Eq. (3) according to

$$\Delta\lambda = 4\pi r_0^2 c n_0^1 Z_{\text{eff}} D \int_{-\infty}^{0} \delta W(\delta)d\delta \Big/ \int_{-\infty}^{\infty} W(\delta)d\delta = 4\pi r_0^2 c n_0^1 Z_{\text{eff}} D(kTK_0/2\pi V_0)^{1/2}. \tag{4}$$

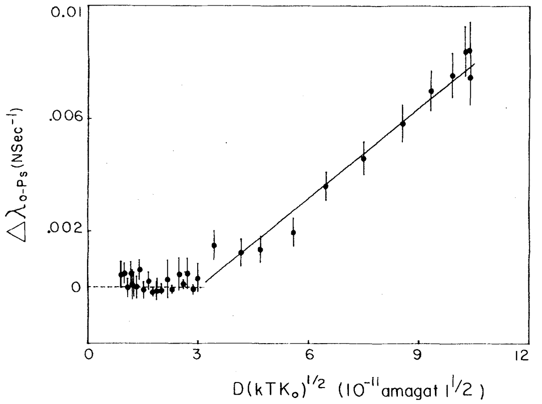

FIG. 4. Deviations $\Delta\lambda$ of the measured orthopositronium annihilation rates vs $D(kTK_0)^{1/2}$ at 377 K. The standard deviations fall within the size of the data points unless otherwise shown. The line represents a weighted least-squares fit to data for $D(kTK_0)^{1/2}$ values corresponding to $20 \leq D \leq 153$ amagats. The dashed line represents the region corresponding to $\Delta\lambda = 0$.

Here $K_0 = -V^{-1}(\partial V/\partial P)_T$ is the isothermal compressibility of the gas and can be obtained from an appropriate equation of state. The measured values of $\Delta\lambda$ are shown as a function of $D(kTK_0)^{1/2}$ in Fig. 3. $\Delta\lambda = 0$ for a range of $D(kTk_0)^{1/2}$ values with an upper limit corresponding to $D = D_1{}^*$. For $D > D_1{}^*$ the behavior predicted by Eq. (3) seems to hold reasonably well for a range of densities from about 11 to 68 amagats at 306.4 K and provides a value of approximately 20 Å for the diameter of the small cell.

At densities higher than 70 amagat at 306.4 K, the observed behavior of $\Delta\lambda$ is certainly complex. In order to compare the application of Eq. (4) to data available for $\lambda_{o\text{-}Ps}$ at temperatures both near and far from the critical temperature of ethane, we have examined the recent $\lambda_{o\text{-}Ps}$ data at the highest temperature, 377 K, investigated in the present series of experiments.[2] The measured values of $\Delta\lambda$ in ethane at 377 K are plotted as a function of $D(kTK_0)^{1/2}$ in Fig. 4. Again, $\Delta\lambda = 0$ for a range of $D(kTK_0)^{1/2}$ values with an upper limit corresponding to a temperature-dependent value of $D_1{}^* \approx 20$ amagats. For a range of densities from about 20 to the highest density of 153 amagats at 377 K, Eq. (4) provides a good fit to the data. This agrees with the results obtained in the case of methane, where $\Delta\lambda$ was observed to vary linearly with $D(kTK_0)^{1/2}$ at 245, 278, and 295 K, far from the critical temperature of methane.

In summary, the measurements reported here support the hypothesis that orthopositronium annihilation rates are sensitive to density fluctuations in ethane gas. The simple first-order approximation model of density fluctuations explains, at least qualitatively, the observed density dependence of $\lambda_{o\text{-}Ps}$ at those densities and temperatures in methane[1] and ethane gases where density fluctuations are small. An understanding of the new features observed in the density dependence of $\lambda_{o\text{-}Ps}$ at high densities in ethane gas at 306.4 K, where the long-range correlations between gas molecules are expected to be important and the density fluctuations are no longer small, awaits improved model calculations.

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