Lifetime of the metastable $^5S^\circ$ state of atomic oxygen*

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(Received 24 September 1973)

The lifetime of the $^5S^\circ$ state of atomic oxygen has been measured as $\tau = 170 \pm 25$ μsec.

Until a few years ago there were no experimental measurements of the lifetime of the aeronomically important metastable $^5S^\circ$ state in oxygen. Then several measurements of the $O(^5S_0)$ lifetime were made. Ott first estimated[1] the lifetime to be $146 \pm {}^{100}_{40}$ μsec using arc-emission techniques. Then Wells and Zipf[2] and Johnson[3] concurrently measured the lifetime using similar time-of-flight (TOF) techniques and found lifetimes of 189 ± 50 and 185 ± 10 μsec, respectively. These results agree well with many-electron theoretical calculations carried out by Nicolaides[4] and are about a factor of 3 shorter than the only previous theoretical calculation, which was done by Garstang.[5] This paper reports yet another measurement of this important lifetime.

This measurement differs from the first preliminary one we reported[2] in that more data were taken and care was exercised to ensure that the detector subtended the same solid angle at both TOF positions. The average result for eleven determinations was 170 ± 25 μsec. This result is consistent with the above-mentioned recent results and does not vary from our earlier measurement in a statistically significant fashion.

The experimental TOF apparatus has been described in detail elsewhere.[6,7] In order to measure the lifetime due to inflight radiative decay in this experiment, the detector was alternately located at a distance of 6.4 and 32.4 cm from the magnetically collimated electron beam (Fig. 1). The pulsed electron beam impinging on the diffuse O_2-gas target dissociatively excited the $^5S^\circ$ state of atomic oxygen. The gun was operated at 20 eV, where only the slower of the two observed metastable peaks[7,8] is generated. Data were recorded automatically using a time-to-amplitude converter and a pulse-height analyzer for detector 1 and a multichannel scale for detector 2. The detector used was a Johnson electron multiplier and worked by secondary-electron ejection off the

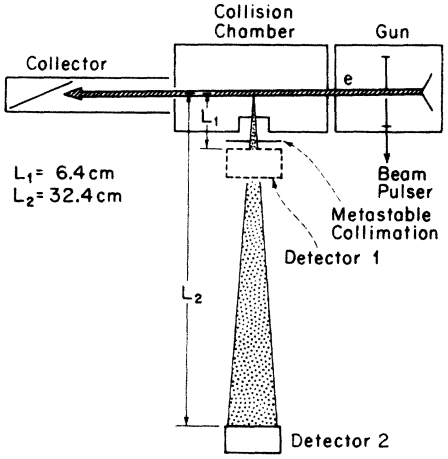

FIG. 1. Experimental apparatus showing the constant solid angle subtended at both positions by the detector. The detector was alternately located at distances L_1 and L_2 from the beam in order to collect data.

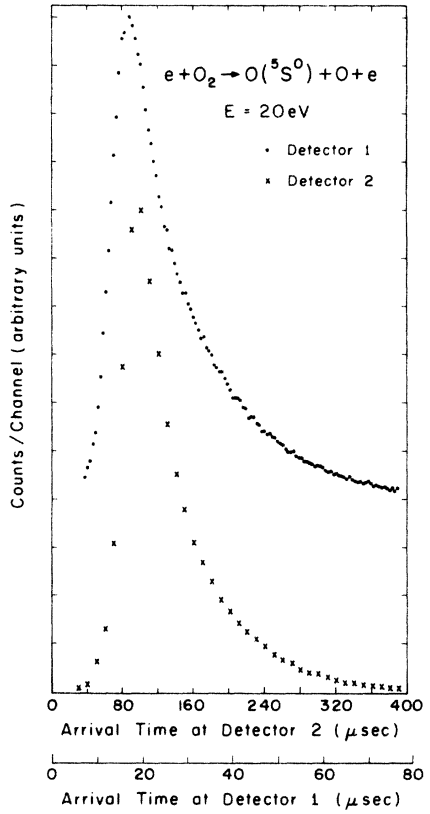

FIG. 2. Typical TOF distributions for dissociatively excited $O(^5S^\circ)$ atoms observed at distances L_1 and L_2 of 6.4 and 32.4 cm, respectively. The data represent about 10 h of collection time. The distributions have been shifted to an equal velocity scale for comparison and division and have been shifted vertically for clarity.

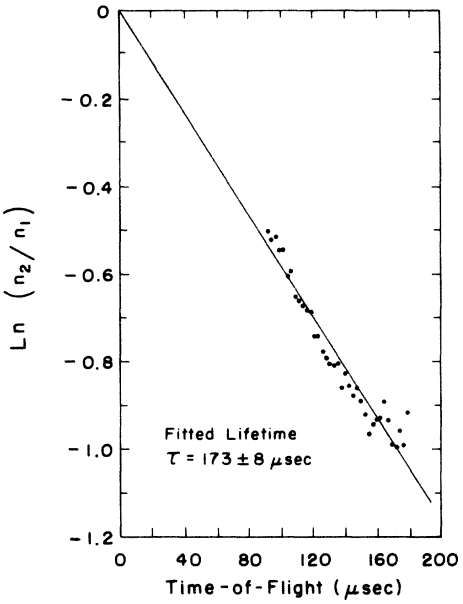

FIG. 3. Typical decay plot. The natural log of the ratio of the equivalent of the velocity distributions at detectors 2 and 1 plotted against the TOF results in a straight line, the slope of which is minus the reciprocal lifetime ($-\tau^{-1}$). The time of flight here is not the arrival time of the excited atom at a detector, but rather the difference of the arrival times at detector 1 and detector 2, $T=(L_2-L_1)/v$. A straight line is fitted through the data points using a least-squares fit, thus yielding the lifetime. The fit for this plot was 173 μsec. The mean of eleven such fits was 170 ± 22 μsec, where the uncertainty represents one standard deviation. Inclusion of systematic errors results in an uncertainty of ±25 μsec.

first Cu–Be–O dynode when struck by a metastable atom. Our surface was insensitive to metastable $O(^1D)$ and $O^1(S)$ atoms. The background was measured before the beam pulse and by assessing the asymptotic value of the tail of the TOF spectra.

Typical TOF spectra are shown in Fig. 2. To deduce the lifetime from this TOF data, one normally transforms the TOF distributions to velocity distributions in order to take their ratio. Equivalently, it is simply possible to shift one TOF spectrum on top of the other to take the ratio. Mathematically, we can express this operation in the following fashion. Taking the distributions detected at L_1 and L_2 to be $n_1'(t)$ and $n_2(t)$, respectively, we can write

$$n_1(t)=n_1'(\alpha t), \tag{1}$$

where $\alpha=L_1/L_2$. Assuming we only lose excited atoms by radiative decay, then

$$n_2(t)=n_1(t)e^{-T/\tau}, \tag{2}$$

where τ is the desired radiative lifetime and T is the time of flight between the two detectors. In this case, we can further write

$$n_2(t)=n_1'(\alpha t)e^{-(1-\alpha)t/\tau}, \tag{3}$$

since $T=(L_2-L_1)/v$, where $v=L_2/t$. From Eq. (2) we see that

$$\ln(n_2/n_1)=-\tau^{-1}T. \tag{4}$$

Figure 3 shows a typical straight-line decay plot, thus demonstrating the validity of Eq. (4) as applied to our data. The slope of the least-squares-fitted straight line is the negative of the reciprocal lifetime τ as seen from Eq. (4). The data were fitted from about the peak in the TOF spectra out to about 10% of the peak in the tail. At longer times the scatter in the data points increased due to poorer statistics; at shorter times distortion was observed that is not fully understood.

Averaging the results from eleven decay plots resulted in a lifetime of $\tau=170\pm25$ μsec. The uncertainty represents one standard deviation for the eleven different measurements and an assessment of the systematic error due to the uncertain-

TABLE I. $O(^5S^\circ)$ lifetime determinations.

Lifetime	Determined by	Method
595 μsec	Garstang	Deduced from theoretical calculation of A_{1355} and A_{1358}
$146\ ^{+100}_{-40}$ μsec	Ott	Deduced from determination of A_{1355} and $(A_{1355})/(A_{1358})$ branching ratio using arc emission techniques
170 ± 25 μsec	This work	Measured using a TOF technique and the slow fragments from dissociative excitation of O_2
185 ± 10 μsec	Johnson	Measured using a TOF technique and the fast fragments from dissociative excitation of O_2
192 μsec	Nicolaides	Theoretical many-electron calculation

ty in the amount of background subtraction. Table I summarizes our present knowledge of the $O(^5S^o)$ lifetime. As can be seen, the recent results are in good agreement.

Particularly noteworthy is the agreement of our result with that of Johnson.[3] The experiments differed in that Johnson used longer drift times and measured the lifetime of the fast dissociative peak, while we measured the lifetime of the slow peak. The identity of excited species in the fast peak are not as well known as in the slow peak.[7] At threshold (14.3 eV) the slow peak is composed of only $O(^5S^o)$ atoms.[7,8] As we go up in energy we can then excite other long-lived states energetically, such as high-lying Rydberg levels in oxygen (O**). The fast peak in the TOF spectra with a threshold of 21 eV is probably originally composed of pure O** atoms[7] which decay, some to $O(^5S^o)$. Thus, the composition of the fast peak changes as a function of time after formation and is probably pure $O(^5S^o)$ after the atoms have drifted 20 cm.[8] Thus, the lifetime measurement of the excited atoms in the fast peak by Johnson,[3] made at distances greater than two meters, was most probably that of $O(^5S^o)$. This is born out by the good agreement between our results.

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*Research supported, in part, by Advanced Research Projects Agency, the Department of Defense, and was monitored by U. S. Army Research Office—Durham, under Contract No. DA-31-124-ARO-D-440, and by the National Aeronautics and Space Administration (NGL39-011-030).

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