Lifetime of the $2p^3\,3s\,^5S$ Metastable State of Oxygen†

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The lifetime of the $2p^3 3s\,^5S$ metastable state of oxygen has been measured using the time-of-flight technique. The velocity distribution of metastable atoms resulting from the dissociation of excited oxygen molecules is sampled and detected at two positions, 1.8 and 6.6 m from the pulsed electron gun used to bombard the ground-state molecules effusing from a source slit. A comparison of the number of metastables within specific velocity intervals at the two detectors determines the number which decay in flight and yields an experimental plot of the number which decay vs time of flight. The lifetime τ is then obtained from the slope $(=-1/\tau)$ of a straight line least-squares fitted to the decay plot. The result is $\tau = 185 \pm 10\ \mu$sec.

The same time-of-flight technique previously employed to measure the lifetimes of the $2\,^1S_0$ metastable state[1] of He, the $c\,^3\Pi_u$ metastable state[2] of H_2, and the $a\,^3\Pi$ metastable state[3] of CO has been used to investigate the lifetime of the $2p^3 3s\,^5S$ metastable state of oxygen. The $3s\,^5S$ state, with an excitation energy of 9.14 eV, is the lowest quintet state of oxygen and therefore is metastable. Its decay to the $2p^4\,^3P$ ground state arises mainly through spin-orbit interaction with the $2p^3 3s\,^3P_2$ level, and the lifetime for this decay has been calculated by Garstang[4] to be about 600 μsec. Two other oxygen metastable states, the $2p^4\,^1D$ at 1.97 eV and the $2p^4\,^1S$ at 4.18 eV, both have an excitation energy less than the work function of our Auger detector and are therefore not observed in this experiment.

A complete description of our apparatus, data-collection scheme, data analysis, and time-of-flight theory has been described previously.[1] The experiment is based on the time-of-flight technique, where an atom or molecule is assumed to leave the metastable state only by radiative decay as it drifts over a 5-m path between two fixed detectors; this assumption demands a very low pressure ($< 10^{-7}$ Torr) in the drift region to minimize scattering losses. Neutral ground-state oxygen molecules effuse from a source slit and are immediately excited by a pulse of magnetically focused electrons. The metastable $O(^5S)$ atoms resulting from the dissociation[5,6] of the excited O_2 molecules are then collimated while passing through three buffer chambers and finally detected at both ends of the 5-m-drift region. The first detector consists of a 60% transmitting copper mesh target. The secondary electrons which are Auger-ejected from the copper surface by the metastable atoms are collected by an EMI electron multiplier. The second detector is a solid copper target and intercepts the transmitted metastable atoms which survive the flight between the two detectors.

The data-taking and timing aspects of the experiment are controlled by an on-line PDP-8 computer. An example of the data collected is shown in Fig. 1 and represents about 10^6 separate collection sweeps during a total collection time of 12 h. The electron

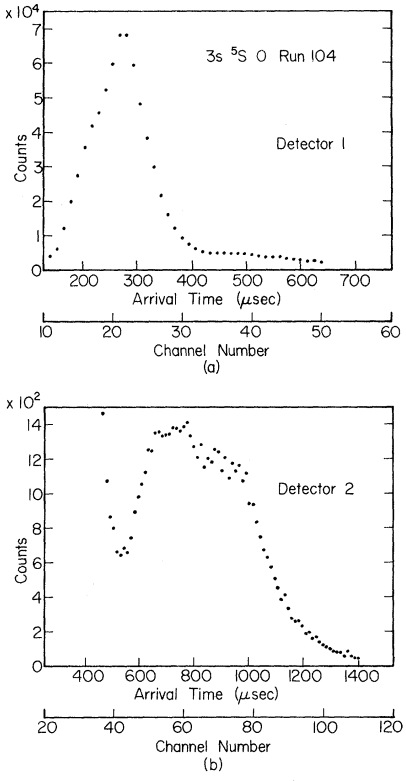

FIG. 1. $O(^5S)$ time-of-flight distributions, representing about 10^6 separate data-collection sweeps. The tail of a large peak appearing in the first few channels of the detector-2 distribution represents neutral atoms or molecules with kinetic energies greater than 20 eV; they result from the acceleration of either O^+ or O_2^+ ions in the electron gun followed by charge exchange in the source chamber region.

gun is pulsed on only during channel 0, and counts are then collected simultaneously at both detectors into 199 channels, not all of which are shown. All channel widths are equal to 12.75 μsec. The amount of background subtraction is obtained from the long tail of the time-of-flight distributions.

The following two expressions represent the number of metastable atoms counted by detectors 1 and 2:

$$n_1(v) = \int_{\text{surf}} \epsilon_1 n_0(v)\, ds_1, \tag{1a}$$

$$n_2(v) = \int_{\text{surf}} \epsilon_2 n_0(v)\, e^{-t/\tau}\, ds_2, \tag{1b}$$

where ϵ is a detector efficiency factor. The initial velocity distribution $n_0(v)$ of the $O(^5S)$ metastable atoms is determined by the dissociation[5,6] of the excited O_2 molecules. The exponential factor in the expression for the number of counts at detector 2 allows for radiative decay of the metastable beam between the two detectors.

After assuring that n_0 is uniform across the beam, the time and velocity dependence can be extracted from the integrals in Eqs. (1). Then, taking a ratio of detector-2 counts to detector-1 counts for atoms of the same velocity, we obtain

$$R(t) \equiv n_2(v)/n_1(v) = C\, e^{-t/\tau}, \tag{2}$$

where t is the time of flight between detectors and C is a constant independent of the initial velocity distribution. The natural logarithm of Eq. (2) gives the equation of a straight line; a straight line is least-squares fitted to the data and the lifetime τ determined from the slope $(= -1/\tau)$.

The initial step in the data analysis involves partitioning the detector-2 data to correspond to the channel width at detector 1 for metastable atoms with the same velocity. The correct partitioning of the data, followed by calculation of the ratio R for metastable atoms of the same velocity, then leads to a plot of $\ln R$ vs time of flight, as shown in Fig. 2. The least-squares straight line is fitted only to data points corresponding to a number of counts greater than 10% of the peak value. In addition, the first few points are omitted from the fit because these points are influenced by the tail of the fast peak shown in Fig. 1(b).

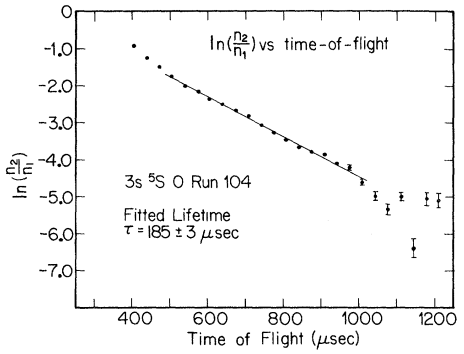

FIG. 2. Decay plot. The ratio of detector-2 to detector-1 metastable atom time-of-flight distributions is a straight line on a logarithmic plot. The measured lifetime τ is obtained from the slope $(= -1/\tau)$ of the least-squares-fitted straight line.

The result for the lifetime of the $2p^3 3s\ ^5S$ metastable state of oxygen is $\tau = 185 \pm 10$ μsec. About half of the error occurs because the electron bombardment region extends for 1 cm along the metastable beam direction, giving an uncertainty in the effective distance to the two detectors. The remaining error results from an uncertainty in the amount of background subtraction at detector 2. All other experimental parameters such as electron gun voltage (100–200 V), source temperature (100 or 300 K), beam flow rate, and drift region pressure have no effect on the measured lifetime.

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[6]The velocity distributions reported in Ref. 5 show two distinct groups of $O(^5S)$ atoms; only the faster group is observed in this experiment since the time of flight, for the slower group is much greater than the measured lifetime of the $O(^5S)$ state.