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Time-resolved two-dimensional imaging of ground-state species using laser-induced fluorescence

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Laser-induced fluorescence has been employed to obtain two-dimensional images of the spatial distribution of a vapor in the ground state with nanosecond resolution. The method has a wide range of potential applications, as it can be used to observe any species which has energy levels accessible to tunable laser systems. In the application we describe, aluminum vapor was observed jetting from spark gap electrodes and diffusing throughout the gap volume.

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INTRODUCTION

In an effort to study the behavior of metal vapor evaporated from spark-gap electrodes a new technique for directly and selectively observing the metal vapor cloud was devised. This technique relies on laser-induced fluorescence (LIF) to make the vapor visible and a SIT vidicon to record the image. The technique provides excellent time resolution determined by the width of the laser pulse and the lifetime of the excited state involved, and is generally applicable for observation of any species with energy levels accessible to tunable laser systems. In contrast to conventional fluorescence techniques, the LIF technique is well suited to the detection of ground-state species.

I. LASER-INDUCED FLUORESCENCE IMAGING

Laser-induced fluorescence is a convenient technique for causing species in the ground or a metastable state to emit light.^{1,2} The process is generally used as a spectroscopic tool, and it can be very effectively used for imaging applications. The LIF process is illustrated in Fig. 1. E_1 , E_2 , and E_3 are three energy levels of the species under consideration. E_1 need not be the ground state, but the strength of the emission will be proportional to the population of E_1 . If the $E_1 \rightarrow E_2$ transition is optically allowed, then a photon of energy $h\nu = E_2 - E_1$ can resonantly pump the system from state E_1 to E_2 . Decay to a lower-lying state will follow and the presence of species in E_2 can be detected by observing the emission resulting from radiative transitions. If in the absence of

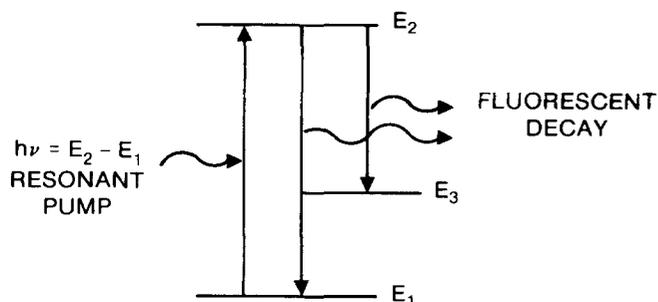


FIG. 1. Energy level diagram of laser-induced fluorescence process.

laser excitation the population of E_2 is low, then for a given laser pump intensity, the intensity of the reemission will be proportional to the population of E_1 during the laser pulse. Temporal resolution is provided, therefore, by the excitation laser, and the detector need not be gated unless it is necessary to reduce interference from background emission.

In this paper we show as an example the application of laser-induced fluorescence imaging (LIFI) to the study of the migration of metal vapor expelled from the aluminum electrodes of a spark gap. Time-resolved images of the ground-state Al vapor cloud obtained with the apparatus clearly show the evolution of the cloud for several hundred microseconds after the spark. The laser was tuned to resonance with the 394.4-nm transition between the ground, $3^2P_{1/2}^0$ state and the $4^2S_{1/2}$ excited state of Al. Reemission from transitions back to the ground state at 394.4 nm and to the $3^2P_{3/2}^0$ state at 396.1 nm was monitored to provide the image.

II. APPARATUS

A schematic diagram of the experimental setup is shown in Fig. 2. The spark gap switches a dc-charged, 20-

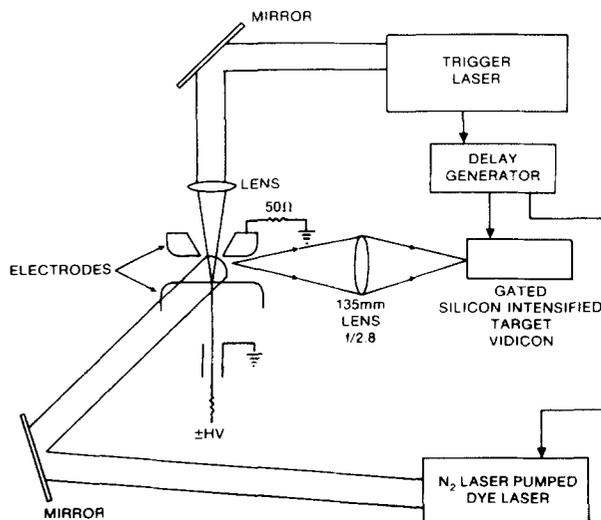


FIG. 2. Experimental apparatus. The pump laser beam passes through the gap perpendicular to the imaging axis.

kV, 50- Ω transmission line into a matched 50- Ω load. The gap is undervolted and is triggered to breakdown by about 50 mJ of focused 1.06- μ radiation from the Nd:YAG laser in a 15-ns pulse. The matched transmission line system produces clean, square-wave pulses of 1 μ s duration and peak current of 200 A. Both the 1.06- μ laser pulse and the spark current are capable of eroding electrode material, and we have obtained photographs of electrode vapor clouds produced by each of these processes.

The basic LIFI system consists of a Molelectron DL-II dye laser pumped by a Molelectron UV400 N2 laser and a PAR 1254 Gated SIT vidicon. The dye laser produces pulses with energy between 10 and 500 μ J, depending on the spectral region, and duration of 8 ns. The active region of the spark gap is imaged onto the face of the vidicon with a 135-mm focal length photographic lens. In order to reduce the interference from the intense emission from the spark, the SIT detector was gated off except during a 50-ns window which contained the pump laser pulse. Further rejection of the unwanted background was provided by placing an interference bandpass filter (400 ± 5 nm) between the lens and the vidicon. The pump laser and the vidicon gate generator are triggered by a multichannel delay generator which is in turn triggered by the laser used to initiate the spark in the spark gap. Varying the delay of the multichannel delay generator provided a convenient means of changing the observation time.

For the pump transition we chose the pump laser was strongly absorbed by the Al vapor. As a result, the beam intensity was depleted in crossing the vapor cloud and LIF was observed only from the surface of the vapor cloud first struck by the beam. Some improvement was obtained by reflecting the pump beam back through the cell, thereby illuminating the cloud from both sides (the beam diameter was much larger than that of the vapor cloud), but an asymmetry is still evident in the pictures we obtained.

III. RESULTS

The migration of the electrode vapor is clearly evident in the sequence of pictures of the vapor cloud starting 5 μ s after the falling edge of the spark shown in Fig. 3. The strong absorption of the pump laser beam provides an edge enhancement effect which clearly defines the boundary of the cloud. The asymmetry in these pictures is due to an unequal illumination of the cloud from each side. Although the edge enhancement seen here may be useful in some applications, the depletion of the pump laser in traversing the vapor cloud precludes a determination of the vapor density in the cloud with this technique. Detuning the pump laser to the edge of the absorption line would reduce the optical density, and hence the pump beam depletion, but self-absorption of the fluorescent emission is likely to still cause problems. A better solution would be to use a more intense pump beam in order to saturate the pump transition throughout the cloud volume.

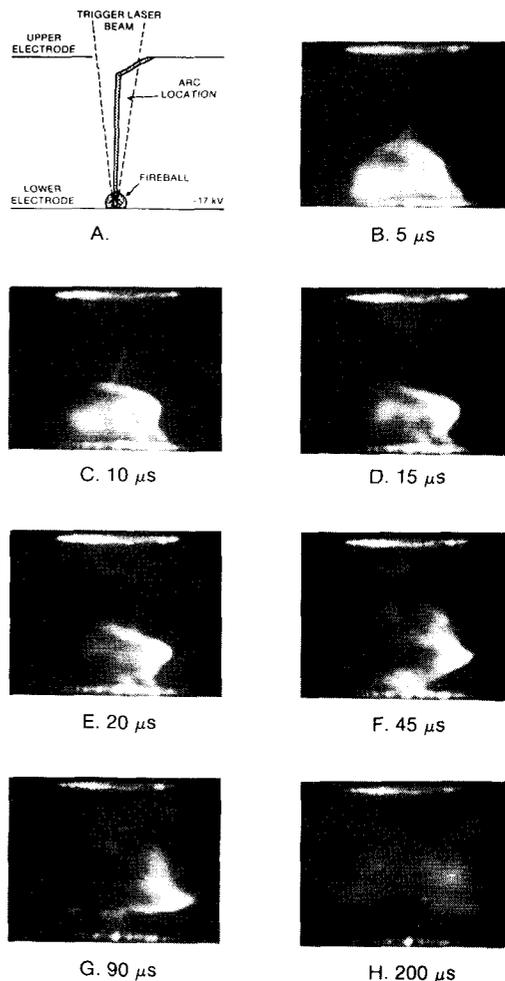


FIG. 3. Time sequence showing the migration of aluminum vapor eroded from the spark gap electrodes by the triggering laser and the arc. (a) Depicted is the location of electrodes in the pictures, (b)-(h) shown are images of aluminum vapor at times as indicated after the arc ended.

DISCUSSION

We have found laser-induced fluorescence imaging to be a very useful and convenient technique for obtaining two-dimensional maps of the presence of ground-state Al vapor in an operating spark gap. The technique should be generally applicable to the observation of any gaseous species with an allowed optical transition accessible to tunable laser sources. The vapors of many metals and other compounds appear to have such transitions. Since the time resolution provided by the technique is determined by the temporal width of the pump laser, the time resolution should be more than adequate for most applications.

¹*Lasers in Chemical Analysis*, edited by G. M. Hieftje, J. C. Travis, and F. E. Lytle (Humana Press, Clifton, NJ 1981), Sec. III.

²V. E. Bondybey and J. H. English, *J. Chem. Phys.* **74**, 6978 (1981).