

# Measurement of the velocity distribution of sputtered Na atoms from NaCl by Doppler shift laser fluorescence

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(Received 22 June 1981; accepted for publication 20 August 1981)

The velocity distribution of sputtered Na atoms from NaCl during 15 keV, Ar<sup>+</sup> bombardment was measured by a Doppler shift laser fluorescence technique. Contrary to other measurements, the sputtered Na atoms were found to have a velocity distribution identical to that of a three-dimensional Maxwell-Boltzmann gas.

PACS numbers: 79.20.Nc, 32.50. + d

It has been shown that ion induced sputtering processes can be classified into the following categories: collisional processes, thermal spike, and thermal evaporation.<sup>1</sup> Thermal evaporation is normally unimportant for targets whose component elements have very low vapor pressures at the target temperature. However, for certain alkali metals and alkali halides, e.g., Na, NaI, NaCl, etc., where either or both atomic constituents possess high vapor pressure at the target temperature the contribution from thermal evaporation is significant, and may be dominant.

However, the exact velocity distribution of the atoms sputtered by this thermal process is still under dispute. The first extensive studies<sup>2</sup> utilized the time of flight technique and consistently showed that the velocity  $v$  distributions  $dS/dv$  of the flux of sputtered alkali atoms had the following form

$$\frac{dS}{dv} \sim v^2 \exp(-mv^2/2kT), \quad (1)$$

where  $m$  is the mass of the alkali atom,  $k$  is the Boltzmann constant, and  $T$  is approximately the target temperature. The intriguing aspect of this  $v$  dependence of the sputtered flux is that Eq. (1) corresponds to the evaporation flux of a two-dimensional gas. Normal evaporation into a three-dimensional gas should have the following  $v$  dependence

$$\frac{dS}{dv} \sim v^3 \exp(-mv^2/2kT). \quad (2)$$

It has been suggested<sup>2</sup> that this loss of one degree of freedom is connected with the limited mobility of the alkali atoms at or near the sample surface. However, recent work<sup>3</sup> by these same authors casts doubt on this measured distribution and suggests a systematic problem with the slit widths used.

These results are also contradicted by the observations of Husinsky *et al.*<sup>4</sup> By use of the Doppler shift laser fluorescence (DSLRF) technique, they reported that during the bombardment of Na halides by 20-keV inert gas ions, the laser fluorescence intensity  $I_f$  from the sputtered Na atoms had the following  $v$  dependence

$$I_f \sim v^3 \exp(-mv^2/2kT). \quad (3)$$

As pointed out by Wright *et al.*, the laser fluorescence is proportional to the number density  $dN/dv$  of the sputtered

atoms in the detected velocity interval.<sup>5</sup> Therefore, the sputtered Na flux would have had the following form

$$\frac{dS}{dv} = \frac{vdN}{dv} \sim v^4 \exp(-mv^2/2kT). \quad (4)$$

In an attempt to unravel these apparently contradicting conclusions, we have studied the energy distributions of sputtered Na atoms from NaCl during 15 keV, Ar<sup>+</sup> bombardment at room temperature with the DSLF technique. However, there are two significant differences from the earlier DSLF experiment.<sup>4</sup> First, in contrast to the high laser intensity used to reach "saturation" in the earlier work, we have used very low (less than 10  $\mu\text{W}/\text{cm}^2$ ) laser intensity to avoid any significant change in the populations of the two ground state levels of the Na atom. At high laser intensity, the number of atoms in the laser beam that have Doppler shifted resonance frequencies within the laser bandwidth can be reduced by fluorescent decay to the other ground state (after which they are no longer in resonance with the laser). The degree of this transfer or "optical pumping" depends on the laser intensity and the length of time spent in the laser beam. Consequently at high laser powers the proportionality between the fluorescence signal and the original number may vary with velocity and thereby distort the measured velocity distribution.

Second, simultaneously with the sputtering DSLF experiment, a similar DSLF scan was performed with a thermal Na vapor cell. This measurement provided the frequency calibration for each scan and allowed the direct determination of the resonance frequency for Na atoms with zero velocity. As is discussed below, the accuracy of the zero velocity point is essential in distinguishing the various velocity distributions.

The experimental arrangement is illustrated schematically in Fig. 1. The tunable dye laser used was a Coherent model 599-21, pumped by a Spectra Physics 166 argon ion laser. The dye laser frequency could be tuned continuously and had a linewidth (FWHM) of less than 20 MHz. The dye laser beam, used to measure the velocity distribution of the sputtered sodium atoms, was mechanically chopped at 18 kHz and then focused on to a 50-m long optical fiber, which transported the dye laser beam from the laser laboratory to the ion accelerator. The output from the fiber was recollimated by a lens to a 5-mm-diam beam which was incident upon the optically polished 5-mm-thick NaCl single-crystal

<sup>a)</sup>This work was partially supported by the Office of Naval Research.

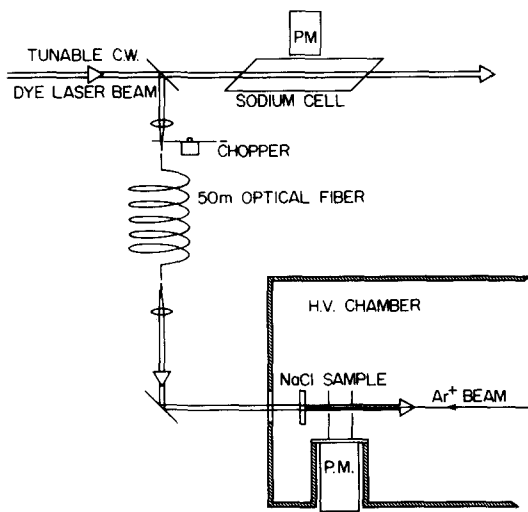


FIG. 1. Schematic diagram of the experiment.

sample. The laser beam passed through the sample, and was centered on the spot bombarded by the 1-mm-diam  $\text{Ar}^+$  beam on the output face. A differentially pumped, mass analyzed, ion gun provided the  $2\text{-}\mu\text{A}$ , 15-keV,  $\text{Ar}^+$  beam for the sputtering at normal incidence. The  $\text{Ar}^+$  beam was chopped at 20 Hz. The chamber pressure was about  $1 \times 10^{-7}$  Torr during the sputtering. The fluorescence from the Na atoms in a 1.5-cm-long region, 1.5 cm downstream from the sample, was monitored at right angles by a photomultiplier. In order to eliminate the interference from the scattered laser light in the chamber, the luminescence of the NaCl crystal during the ion bombardment, and the light emission from the sputtered excited atoms, a dual lock-in scheme was used. The photomultiplier output was first demodulated by a PAR HR-8 lock-in amplifier with a Q of 5 at the laser chopping frequency of 18 kHz. The mixer output was fed directly without filtering into a second lock-in amplifier tuned to the ion beam chopping frequency of 20 Hz for the final stage of demodulation. The output of this lock-in went to one pen of a dual pen chart recorder, the other pen of which simultaneously recorded the fluorescence from the sodium vapor cell.

This sodium vapor cell contained no buffer gas in order to ensure that there was no pressure shift of the frequency of the sodium  $D_2$  line at 5890 Å. Typically, the cell pressure was less than  $10^{-5}$  Torr and the sodium atomic number density was  $1 \times 10^9$ . The measured cell temperature was 130 °C. As discussed earlier,<sup>6</sup> the intensity of the 5-mm-diam laser beam traversing the cell was less than  $0.1 \mu\text{W}/\text{cm}^2$  to eliminate any optical pumping effects. The fluorescence from the Na vapor was monitored by a cooled photomultiplier (PM) connected to one pen of the dual pen recorder while the laser frequency was slowly swept across the sodium  $D_2$  line. For our dilute vapor conditions the shape of the resulting fluorescence versus frequency curve is the same as that for the absorption coefficient of the Doppler broadened line. Thus, this curve provides the frequency calibration and the zero velocity point.

Typical measurements are shown in Fig. 2 together

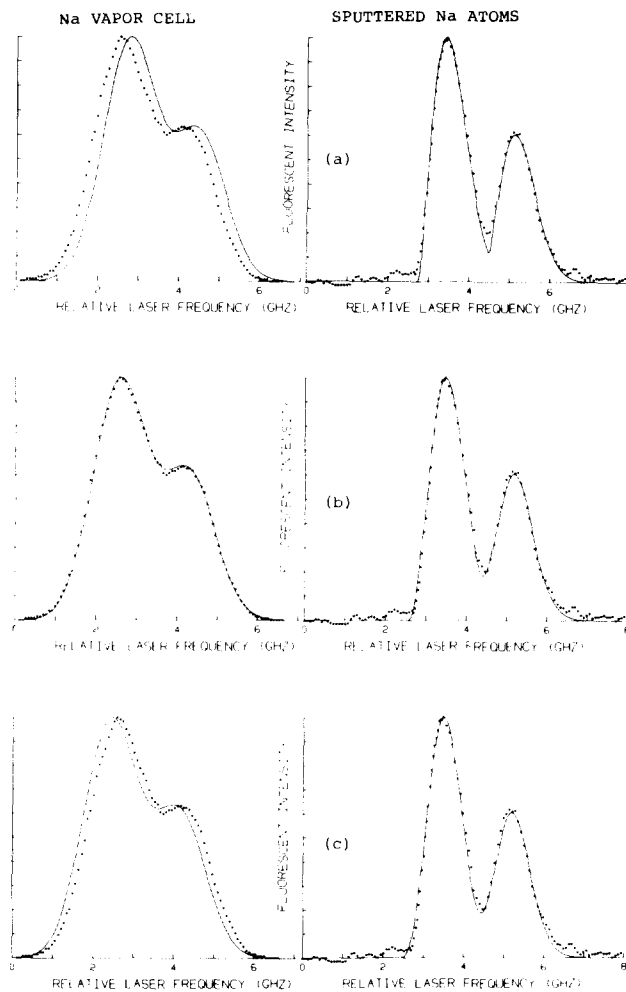


FIG. 2. Comparison of theory (solid lines) with experiment (dots) for the Doppler shift laser fluorescence (DSLRF) scans of the Na vapor cell and the sputtered Na atoms. The fluorescence is proportional to the number density of atoms with velocity  $v$  within the velocity interval  $\Delta v$  determined by the laser bandwidth of 20 MHz. A 1-GHz Doppler shift corresponds to a  $5.89 \times 10^4$  cm/sec velocity along the laser beam. The frequency for the zero velocity Na atoms was numerically determined from the best fit of the DSLRF scan to the following number density distributions for the sputtered atoms. (a)  $v \exp(-mv^2/2kT)$ , (b)  $v^2 \exp(-mv^2/2kT)$ , and (c)  $v^3 \exp(-mv^2/2kT)$ .

with comparisons of three different velocity distribution functions for the sputtered atoms. The curve on the left is the fluorescence versus frequency measurement for the Na vapor cell while the curve on the right is the same measurement for the sputtered sodium atoms. The measurements are indicated by the dots which have been digitized from the dual pen recorder traces. The data is the same for Figs. 2(a)–2(c). The double humped shape of both curves is due to the fact that the ground state of the sodium atom is split into two substates separated by 1.77 GHz, thereby causing the appearance of two overlapping lines.<sup>7</sup> The ratio of the strength of these two lines is a sensitive indicator of optical pumping effects. When optical pumping occurs the ratio changes from 0.6 to smaller values. As can be seen the observed ratios indicate optical pumping can be neglected here. The two fluorescence curves have different shapes and appear at different

frequencies. This occurs because the fluorescence from the sodium vapor cell is a measure of the number density of atoms within the interval of velocity components along the laser beam determined by the frequency and bandwidth of the laser. However, compared to the vapor cell the sputtered atoms must in addition be within the solid angle determined by the laser beam area and distance from the sample in order to be detected. Also for the sputtering case the Doppler shift is only in one direction.

One of the most surprising features of our theoretical analysis is that the fluorescence from the sputtered atoms can be well fit by any one of the three distributions for the number density  $dN/dv$  of atoms with velocity  $v$ : (a)  $v \exp(-mv^2/2kT)$ , (b)  $v^2 \exp(-mv^2/2kT)$ , and (c)  $v^3 \exp(-mv^2/2kT)$ . Multiplication of these expressions by  $v$  gives the atomic flux velocity distributions  $dS/dv$  of Eqs. (1), (2), and (4), respectively. For the sputtered atoms the numerical fit involved two adjustable parameters, the relative frequency  $\omega_0$  corresponding to  $v = 0$  (Ref. 9), and the temperature  $T$ . The values obtained for best fit were: (a)  $\omega_0 = 2.81$  GHz,  $T = 380$  °K, (b)  $\omega_0 = 2.60$  GHz,  $T = 370$  °K, and (c)  $\omega_0 = 2.43$  GHz,  $T = 350$  °K. If only the right-hand curves are considered, all three fits are excellent. The evident experimental difficulty in separating these three distributions, if only the sputtered atoms are measured, accounts, we believe, for the previous conflicting observations of a distribution equivalent to (a), and the observation of (c). However, if the frequencies corresponding to  $v = 0$  for (a), (b), and (c) are then used in a numerical fit of the fluorescence from the sodium vapor cell to the Gaussian distribution  $\exp(-mv^2/2kT)$ , the indicated obvious differences are ob-

tained. Clearly, only the  $v = 0$  parameter as determined by the (b) distribution is consistent with the reference measurement. Therefore, the velocity distribution  $dS/dv$  of the flux of the sputtered atoms is proportional to  $v^3 \exp(-mv^2/2kT)$ , which is exactly the same as that characteristic of the effusive flow of a perfect three dimensional gas through an aperture.

We would like to acknowledge useful discussions with A. H. Nethercot, W. Reuter, and A. C. Tam. In addition, time on the ion beam machine was generously provided by W. Reuter.

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<sup>6</sup>D. Grischkowsky, Appl. Phys. Lett. **36**, 711 (1980).

<sup>7</sup>Each peak is actually an overlap of three lines due to the smaller hyperfine splitting of the  $3^2P_{3/2}$  excited state.<sup>\*</sup>

<sup>8</sup>W. M. Fairbank, T. W. Hänsch, and A. L. Schawlow, J.O.S.A. **65**, 199 (1975).

<sup>9</sup>Each theoretical distribution is the sum of six curves, corresponding to the six hyperfine lines and weighted by their strengths.  $\omega_0$  refers to the strongest line, ( $F' = 3 - F = 2$ ), which fixes the entire comb.<sup>\*</sup>