GENERAL PHYSICS

I. MOLECULAR BEAMS*

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RESEARCH OBJECTIVES AND SUMMARY OF RESEARCH

We have been using molecular-beam techniques to study superfluid liquid helium and other systems at low temperature. The following helium beam experiments are in progress.

1. Measurement of the cross section of ^{4}helium atoms for collision with both ^{4}helium and ^{3}helium at low temperatures (D. E. Oates).

2. Measurement of the velocity distribution of atoms evaporating from helium II at low temperatures (J. W. McWane and R. F. Tinker).

3. Studies of evaporated atoms resulting from heat pulses introduced in the liquid (R. F. Tinker).

4. Studies of diffusion of ^{3}helium in ^{4}helium by observation of evaporating atoms (G. A. Herzlinger).

5. Investigation of motion of ions in helium at low current densities (W. B. Davis, with Dr. C. E. Chase of the Francis Bitter National Magnet Laboratory).

6. Studies of evaporation from superfluid helium films (J. R. Clow).

We are continuing and accelerating our work in various kinds of microscopy which includes the computer design of improved electron microscope lenses (D. S. Ofsevit) and their experimental realization (E. H. Jacobsen). We are also developing a scanning neutral molecule "microscope," using water molecules with which we plan to study various biological systems (J. C. Weaver).

J. G. King, J. R. Zacharias

A. IONS AT LOW DENSITIES IN He II

Many experiments on properties of ions in liquid He II have been carried out. In every case, however, the ions were detected with the use of standard electrometer techniques requiring currents in range 10^{-12}-10^{-14} A. Qualitative calculations suggest that space-charge effects may be important in some cases, especially at the lowest temperatures.

Application of electron multiplier techniques makes it possible to detect single ions,

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thereby increasing the sensitivity by a factor greater than 10^4. In order to accomplish this, the ions must be extracted from the liquid and accelerated to a high velocity in a near vacuum.

An apparatus is being constructed to test this procedure. Once its feasibility has been demonstrated, a variety of experiments can be carried out; in particular, we hope to study the "neutral excitations" recently reported by Surko and Reif.[1]

W. B. Davis, C. E. Chase, J. G. King

(Dr. C. E. Chase is with the Francis Bitter National Magnet Laboratory.)

References

1. C. M. Surko and F. Reif, Phys. Rev. 175, 229 (1968).

B. VELOCITY DISTRIBUTION OF ATOMS EVAPORATING FROM SUPERFLUID HELIUM

1. Introduction

In 1966, Johnston and King[1] reported the first measurements of the velocity distribution of atoms evaporating from the surface of bulk liquid helium. They found the distribution to be the same as a Maxwell-Boltzmann gas of a temperature substantially higher than the liquid source. For a liquid-source temperature of 0.6°K the distribution was characteristic of a gas of 1.6°K. Since the publication of their results, several attempts have been made to understand the helium evaporation process,[2-7] and experiments have been suggested to test their hypotheses.

This report discusses the progress on experiments to confirm the measurements of Johnston and King and to extend them over a wider range of temperatures. Several other experiments are discussed that can be performed with the same apparatus.

2. Description of the Experiment

The experimental apparatus for measuring the velocity distribution is shown schematically in Fig. I-1. It uses the chopped-beam, time-of-flight technique of Johnston and King, but differs in one important respect. In this apparatus all of the atomic-beam measuring components are at the temperature of liquid helium.

The He^4 source chamber a is cooled by the He^3 refrigeration chamber b. This is a one-shot, nonrecirculating He^3 system whose temperature can be varied down to ~0.3°K. The source chamber is supplied by a length of 0.008 in. ID capillary c from a scintered copper condensation chamber d located in the He^4 which is filled by a larger capillary from room temperature. The capillary c is thermally anchored to the

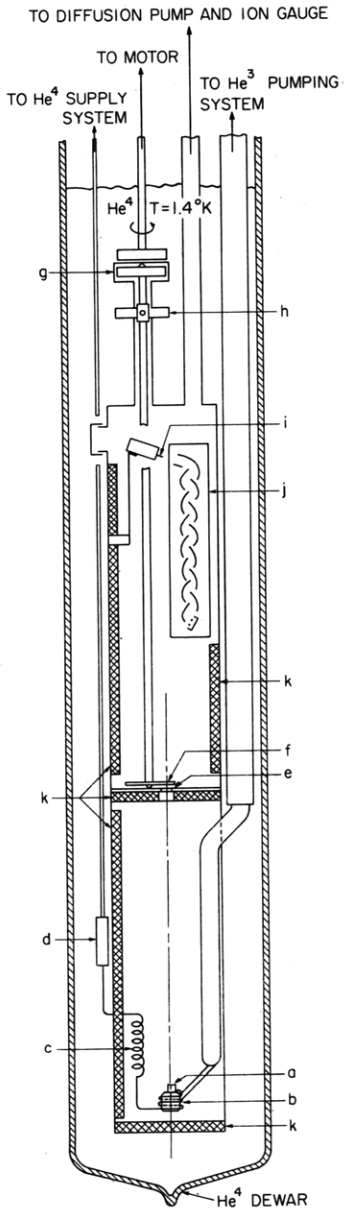

Fig. I-1. Apparatus for measuring velocity distributions of atoms evaporating from superfluid helium.

refrigerator by soldering several turns of it around the outside of the cooling chamber before it enters the source chamber. The beam passes through the collimating slit e and is chopped by the disc f. The chopper is driven by a motor at room temperature, but is thermally isolated from it by the magnetic coupling g. After passing through the drift space, the chopped beam is detected by the tungsten needle i, with the use of the field-ionization technique.[8] This operation drives the ionized atoms into the electron multiplier j, which has an amplification of approximately 10^6. The resulting pulse is then taken to room temperature where it is further amplified.

The detector needle can be mechanically swept in the two orthogonal dimensions perpendicular to the beam by two bellows pusher assemblies (not shown). This permits proper positioning of the detector by analysis of the beam profiles. A trigger pulse marking the opening of the beam is obtained by shining a light through a hole in the

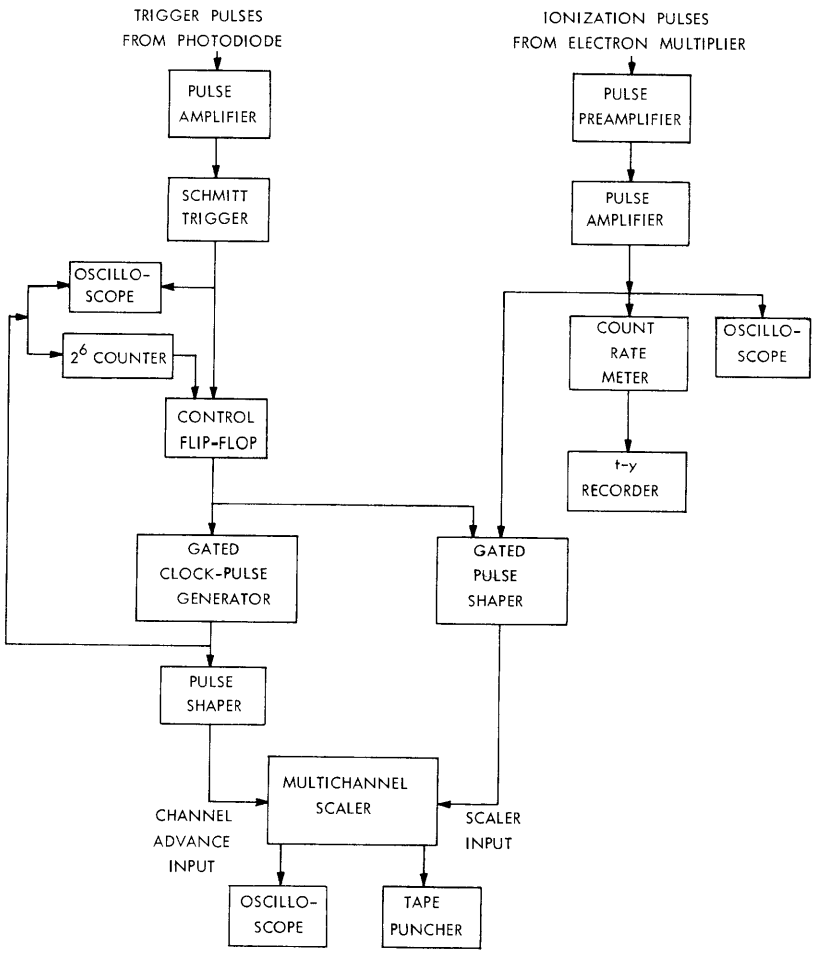

Fig. I-2. Detector electronics.

chopper shaft and detecting it with a photodiode at h. The vacuum in the experiment, which is necessary for achieving long mean-free paths and low background count levels, is provided entirely by the cryosorptive pumping properties of molecular sieve zeolite. This is in the form of pellets that are held to the cold walls k by retaining screens.

The arrival-time spectrum is analyzed by the electronics system shown schematically in Fig. I-2. The ionization pulses are amplified, their rate monitored continuously on a recorder, and fed into a gated pulse shaper before going to a multichannel scaler. The scaler is activated from the chopper trigger pulse. The live time per channel may be varied, as well as the initiation time of the scaler relative to the chopper trigger, so that the spectrum can be analyzed in some detail. The stored spectrum is printed out on paper tape, and this is later fed into a computer for graphing and analysis.

3. Performance Tests

All components of the apparatus perform to expectation, except for the pumping ability of the zeolite. The He^3 refrigerator achieves a low temperature of 0.296°K and can maintain that for more than 10 hours under no-load conditions. The dominant heat load to the refrigerator is thermal conduction to the He^4 bath during the filling process by the superfluid helium in the fill capillary. Several fillings can be made on a single charge of He^3.

The tungsten needle ionizer and electron multiplier amplifier perform reliably at the low temperatures in detecting the helium atoms. Beam profiles that agree with predictions have been made with the chopper locked open, by sweeping the needle across the beam parallel and perpendicular to the mouth of the electron multiplier. Beam profiles perpendicular to the electron multiplier are modified by an increasing detector efficiency of the electron multiplier as the needle is moved toward its mouth. The ionized atoms come off the needle in a cone and the electron multiplier subtends a larger part of this cone, the closer the needle. The geometric tip radius of the needle can be determined in situ from the Fowler-Nordheim plot of electron current vs negative needle voltage. These plots yield typical tip radii of several hundred Å. Operated at a positive potential the helium ionization count rate vs voltage is similar to that reported by Southon and Brandon,[9] and the needle is operated in the "working range" in which $N \propto V^n$, where N is the number of ionized atoms, V is the needle voltage (≈12-20 kV), and n is of the order of 3. This regime has been analyzed theoretically by Southon[10] who finds the supply rate to the needle to be dominated by a dipole attraction of the incident atoms. This leads to an ionization probability proportional to $\frac{1}{2} \alpha E^2 / \frac{1}{2} mv^2$, where α is the atom's polarizability, $1/2\ mv^2$ its kinetic energy, and E the electric field. Such a velocity dependence of the ionization probability must be determined in order to extract meaningful velocity distributions from the data. Johnston[11] saw no such velocity dependence

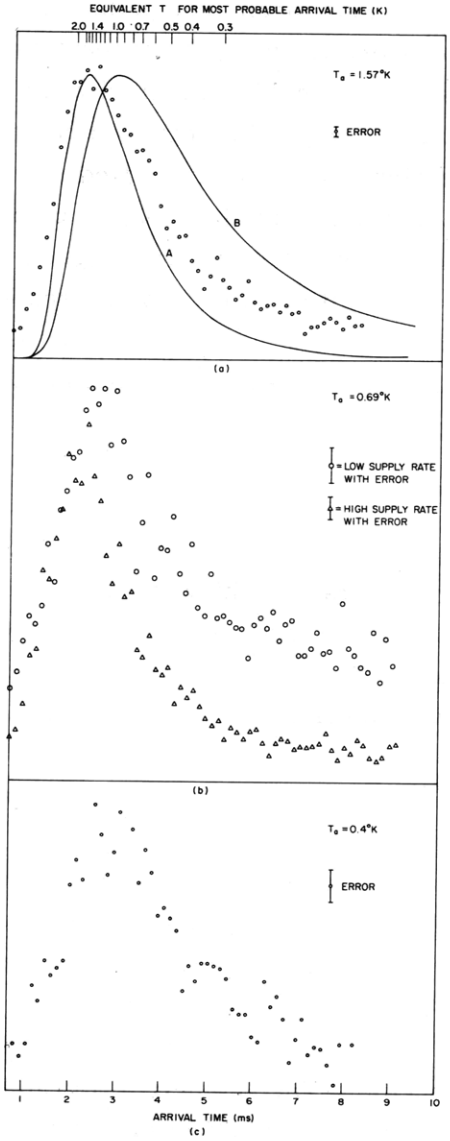

Fig. I-3. (a) Curve 1: Experimental arrival time data from gas source at 1.57 °K.
Curve 2: Arrival time from Maxwell-Boltzmann theory (uncorrected for finite gate function and slow atom scattering).
Curve 3: Same as curve 2 but modified by $1/v^2$.
(b) Arrival time data from non-gas source at 0.69 °K.
(c) Arrival time data from non-gas source at 0.4 °K.

from his analysis of gas distribution at 4.2°K. The current data support that result. Figure I-3 shows time-of-flight data for a gas in the source chamber at 1.5°K. Curve A is that which would be expected from a 1.5°K Maxwell-Boltzmann gas and a velocity-independent ionization probability. Curve B shows an ionization probability proportional to $1/v^2$.

The zeolite used to maintain the low background pressure necessary for the experiment has not yet worked sufficiently well to get velocity distribution from bulk liquid. The requirements on the zeolite are that it maintain a pressure of the order of 10^{-6} T anywhere the beam is (a mean-free path ≈100 cm), but 10^{-9} T in the detecting chamber (a background count rate of ≈1000 counts/sec). When the system is first cooled to liquid-helium temperatures and before any helium has been admitted to the source chamber, the background count rate is less than 0.1 count/sec (≈10^{-13} T). With the source on, the mean-free path criterion is easily met by the zeolite but under the best circumstances the background count level criterion is met for input rates of only 4.10^{17} atoms/sec. This is not sufficient to maintain bulk liquid in the source chamber, since film flow out of the chamber alone supplies 4.10^{18} atoms/sec. Thus velocity distributions have only been obtained for gas and small quantities of liquid/film in the source. This will be discussed further.

The pumping speed of the zeolite has therefore not equalled that reported by Johnston.[11] He estimated an average sticking probability of $1\text{-}10^{-10}$, while in this apparatus it is substantially less for the same zeolite configuration and equivalent input rates. The poor pumping speed has been attributed to poor thermal contact of the zeolite to the 1.4°K bath and thus its inability to carry away the heat of adsorption. Substantial adsorption occurs only when the temperature of the adsorber is near the condensation temperature of the material to be adsorbed and improves as the temperature is reduced.[12] Rough estimates of the thermal conductivity at these temperatures and helium input rates indicate that temperature rises of several tenths of a degree are possible for our configuration, and this would severely alter the zeolite as an effective pump.

Several steps are planned to improve the vacuum in the detecting chamber. The diameter of the source chamber will be made smaller to reduce the film flow supply rate to the background. Furthermore, an investigation is under way to determine an effective means of bonding zeolite directly to the apparatus walls as reported by Stern et al.[13] By mixing small copper wires into the zeolite, substantially better thermal conductivity should be achieved. Finally, the technique of differential pumping will be used, in which the beam must pass through several successive zeolite-coated pumping chambers before reaching the detecting chamber. This improves the collimation and reduces the background supply.

4. Results

Because of the low pumping speed of the zeolite, it has not been possible to obtain velocity distributions of atoms evaporating from the bulk liquid. Gas distributions have been obtained, however, for several temperatures, as well as distributions from non-gas sources. The latter were taken with the capillary supply pressure greater than that necessary to condense liquid in the capillary but less than that necessary to fill the chamber with liquid. Thus, small quantities of liquid were effusing into the chamber and evaporating without accumulating enough for positive identification as bulk liquid. This probably results in distributions that are combinations of liquid, gas and film. These distributions appeared consistently hotter than the actual source temperature Ta (as determined from the ^{3}He vapor pressure in the refrigeration chamber), in confirmation of the results of Johnston and King. For this preliminary analysis the distributions were assumed to be Maxwell-Boltzmann and the apparent temperature T^* of the source was determined by the position of the most probable arrival time, given by $t = \sqrt{\frac{2}{5}} \sqrt{\frac{2kT^*}{m}}$. T^* was seen to change during the course of a single run while the chamber temperature Ta remained constant. Figure I-3b shows two distributions taken at 0.69 °K in which only the supply rate to the source chamber had changed. For the higher supply rate $T^* = 1.7$°K, while for the lower $T^* = 1.2$°K. Figure I-3c is a distribution taken at 0.4°K showing a T^* of 1.0°K, but is significantly broader than a pure Maxwell-Boltzmann at that temperature.

These preliminary data indicate that T^* is substantially higher than Ta over a range of temperature from 0.3°K to 0.8°K. More quantitative data should be realized from experiments now in progress.

5. Theory and Future Experiments

Theories capable of accounting for the Johnston and King observations have been suggested. We shall summarize them in order of their appearance.

John G. King[14] has pointed out the extreme sensitivity of the evaporating atom velocity distribution on the details of the potential near the surface. For instance, there could be a potential barrier caused by the increased density of atoms at the Helium surface. This potential barrier could easily be of such height and width as to strongly attenuate low-energy evaporation, and perhaps also, through Ramsauer tunneling, enhance evaporation of atoms possessing certain energies. This energy dependence would strongly modify any of the following theories which explicitly assume no energy dependence in the creation of free atoms. A wide spectrum of effective temperatures, velocity distributions, and rates could be approximated by proper choice of a few barrier parameters.

Franz Mohling[2] has pointed out that evaporating rotons could make the evaporating velocity distribution appear warmer.

A theory relating evaporation directly to the quantum state of the bulk Helium has been put forward by J. W. Gadzuk.[3] He notes that, because of the microscopic occupancy of the ground state, fluctuation currents are significant in superfluid Helium. He proposes that the fluctuation current actually is the evaporating current and develops an expression for it in the Bogolubov approximation. He predicts a Maxwellian velocity distribution dominated by an $\exp(-\mu/2T)$ temperature dependence (μ is the chemical potential, 7.18°K).

R. F. Tinker has drawn attention to the question of momentum conservation in the creation of evaporating atoms from excitations.[4] If only momentum perpendicular to the surface is conserved, the only effect of the dispersion relation $\epsilon(p)$ on evaporation is the number of values of its inverse $p(\epsilon)$. A Maxwellian distribution of the expected temperature is generated by the phonons up to the maximum at 13°K, and the tail of another Maxwellian by the rotons above Δ. This results in a characteristic distribution around 0.6°K qualitatively like Fig. I-4. The sharp peak that is due to the tail of the

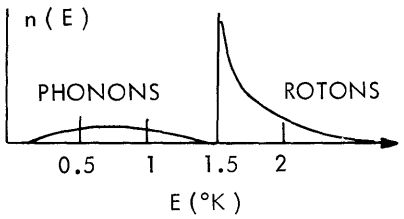

Fig. I-4. Possible velocity distribution for a source at 0.6 °K. The height and shape of the second peak, resulting from evaporating rotons, is arbitrary.

roton distribution starts at $\Delta - \mu = 1.5$°K.

If momentum is conserved upon evaporation, only three simple processes involving phonons can contribute significantly. Rotons cannot easily get rid of their relatively large momentum; extremely unlikely and energetic collisions are required (see Fig. I-5). The possible processes are: direct phonon conversion near $\epsilon_x = 8.53$°K, where the phonon and free-particle curves cross; two-phonon decay that is possible only for total energies $\epsilon < \epsilon_x$; and phonon to roton conversion with emission of a soft phonon that is possible only for incident energies $\epsilon < \epsilon_x$. This last process, if it dominated the evaporation, could account for the Johnston and King results. It would produce a distribution sharper than Maxwellian with an effective temperature always above $\epsilon_x - \mu = 1.35$°K.

Widom has developed a tunneling theory analogous to superconduction tunneling.[5] He points out that detailed balance relates evaporation and adsorption from the vapor. He states that the results require that below 0.7°K the adsorption probability must be $\lambda e^{-\epsilon/\tau}$, where $\tau \sim 1$°K and consequently the effective temperature $T^* = \frac{T - \tau}{T\tau}$.

David Hyman, Marlan Scully, and Allan Widom[6] have considered various evaporation

processes in detail. They develop a lifetime-broadened roton evaporation theory that peaks sharply at $\Delta - \mu$. They believe that this dominates the evaporation process in part because of the large density of states available to rotons. Phonons evaporating would result in a low broad distribution peaked at 0.4°K in their model. Multi-excitation processes are assumed to account for the remainder of high-energy evaporating atoms.

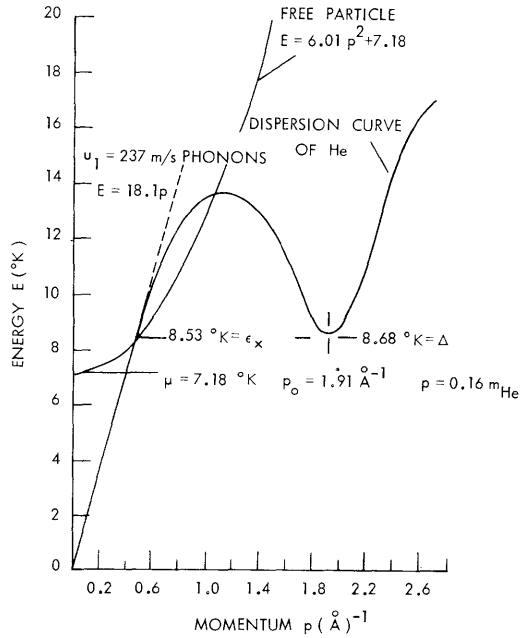

Fig. I-5.

Helium quasi-particle dispersion curve below 1 °K with the free-particle curve displaced μ (latent heat per atom) above zero.

Phillip W. Anderson[7] states that the Johnston-King results virtually require that the single-roton process dominate evaporation, and gives very general phase-space arguments to support this contention. He also posits less favorable phonon evaporation, peaking around ϵ_x or lower. These two sources result in an expected evaporation distribution similiar to Fig. I-4.

Further experiments are needed to choose between the theories and to shed more light on the details of the evaporation process. We expect that the present experiment will yield accurate velocity distributions, their effective temperatures, and the temperature dependence of the total evaporation rate over temperatures from 0.3°K to 0.8°K. From these data we should be able to determine which processes dominate evaporation in this range.

In order to gain more detailed information about the dominating processes, a further experiment would involve pulsing the Helium source with heat. For small, relatively warm sources this will yield information about any delay and conversion times in the evaporation mechanism greater than 100 μs. For large, cold sources the heat pulse generates essentially free excitations that travel to the surface at their various group

velocities. Consequently, the evaporation probabilities of rotons and phonons can be found by determining the rates of evaporation and velocity distributions as functions of time after a heat pulse. More precise information on excitation evaporation probabilities could be obtained by generating monochromatic excitations from a tunable microwave source in the 200-GHz range. These two techniques are capable of determining in detail what excitation-evaporating atom interactions are involved in evaporation, including surface modes or ripplons.

Another proposed series of experiments involves velocity analysis of evaporation from films of varying thickness and orientation. To the extent that evaporation from bulk Helium is understood, this could yield important new information about the superfluid film. For instance, if rotons dominate evaporation, Δ as a function of film thickness could be found as Hyman, et al. suggest. Evaporation measurements from curved films covering spheres or wires could indicate whether surface potentials or ripplons were important in the evaporation process.

Measurements of evaporation from ^{3}He and dilute solutions of ^{3}He in ^{4}He will be attempted. The existence of temperature enhancement of the ^{3}He evaporation distribution would almost certainly rule out theories relying on properties of the phonon-roton dispersion curve. Values of parameters in the dilute ^{3}He dispersion curve might result from these measurements.

J. W. McWane, R. F. Tinker

References

1. W. D. Johnston, Jr. and J. G. King, Phys. Rev. Letters 16, 1191 (1966).
2. F. Mohling, Private communication, 1969.
3. J. W. Gadzuk, Phys. Letters 27A, 23 (1968).
4. R. F. Tinker, Quarterly Progress Report No. 92, Research Laboratory of Electronics, M.I.T., January 15, 1969, pp. 1-10.
5. A. Widom, Phys. Letters 29A, 96 (1969).
6. D. S. Hyman, M. O. Scully, and A. Widom (to appear in Phys. Rev.).
7. P. W. Anderson, Phys. Letters 29A, 563 (1969).
8. W. D. Johnston and J. G. King, Rev. Sci. Instr. 37, 475 (1966).
9. M. J. Southon and D. G. Brandon, Phil. Mag. 8, 579 (1963).
10. M. J. Southon, in Field-Ion Microscopy, J. J. Hren and S. Ranganathan (eds.) (Pleuum Press, New York, 1968, Chap. 2).
11. W. D. Johnston, Ph.D. Thesis, Department of Physics, M.I.T., February 1966.
12. C. E. Grenier and S. A. Stern, J. Vac. Sci. Technol. 3, 334 (1966).
13. S. A. Stern, R. A. Hemstreet, and D. M. Ruttenbur, J. Vac. Sci. Technol. 3, 99 (1966).
14. J. G. King, Private communication, 1969.