# 6. SOURCES OF METASTABLE ATOMS AND MOLECULES

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## 6.1 Introduction

This chapter reviews the various techniques used to produce beams of neutral metastable atoms and molecules. Metastable excited states cannot decay quickly to a state of lower energy because the necessary transition is forbidden by dipole and/or spin–flip selection rules. Because of their large internal energy ($\sim$10 eV) and long natural lifetimes ($\gtrsim 10^{-5}$ s), such species play an important role in a variety of situations involving planetary atmospheres, discharges, plasmas, and vapor deposition techniques [1–4] and are essential in many basic studies of atomic collisions [5–19], laser cooling [20, 21], atomic spectroscopy and interferometry [21–23], nuclear physics [24, 25], and quantum electrodynamics [26, 27]. They are also valuable surface-specific probes of condensed matter phenomena [28–31]. We also consider methods of metastable beam modification and characterization; the detection of metastables is the subject of Chapter 11. Metastable sources have been discussed by a number of other authors [32–37]. Moreover, we have found several papers reporting the details of specific sources [8, 9, 11, 18, 26, 28, 38–48] to be particularly comprehensive and useful.

Metastable sources belong generally to one of five categories:

1. Electron-beam bombardment
2. Discharge
3. Charge transfer
4. Optical pumping
5. Thermal

The primary metastable production mechanism in discharge and electron bombardment sources is the impact excitation of ground states by electrons to metastable states. In discharge sources, this occurs in the active plasma of an electrical discharge or arc. Electron bombardment sources, on the other hand, use a well-defined electron beam to excite an effusive or supersonic beam of ground state atoms or molecules. Some sources involve two discharge regions or extremely high electron beam fluxes and are best viewed as hybrids between the two categories.



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In charge-transfer sources, a fast beam of ions is partially neutralized in a target cell, with both ground and excited states being produced. Most excited states which are not metastable decay in a drift region or can be field-ionized, leaving primarily ground and metastable states remaining in addition to the residual ion beam. Optical pumping sources are similar conceptually to those involving electron-beam bombardment, except they use photons to excite the ground state to a level above the desired metastable state. Subsequent decay produces the metastable species. This two-step process is unavoidable with photon excitation because dipole selection rules must be obeyed, whereas electron bombardment excitation is not subject to these rules, and metastable excitation can proceed directly. Thermal sources are based on the fact that a number of elements, including Tl and Ga, have metastable states very close in energy to their ground states. When they are heated in an oven to temperatures sufficient to produce high vapor pressures, the Boltzmann factor for the upper level is large enough to yield an effusive beam rich in metastables.

Sources of a wide variety of metastable atoms and molecules have been developed to date. A representative number of these, listed by the respective metastable species they produce, are listed in Table I. (Table I does not include reference to metastable experiments in which little or no source information is given. Virtually every element in the periodic table and most molecules have metastable states; see references [32–37] and references therein for reports on work with other metastable species.) The large majority of sources has been developed for the production of the metastable states of hydrogen ($2^2S_{1/2}$), helium ($2^1S_0$ or $2^3S_1$), and the heavy noble gases neon, argon, krypton, and xenon ($n^3P_0$ and $n^3P_2$, where $n = 3 \cdots 6$ for Ne through Xe, respectively). Metastable fluxes, as reported in the various references, are given in Table I to provide a general comparison of relative source performance. The reader is cautioned, however, that such fluxes are determined in a variety of ways and, on occasion, have proven to be unreliable. This issue is taken up in Section 6.7 and also in Chapter 11.

Each of the source types listed above has advantages and disadvantages, both generically and with regard to the production of specific metastable species. Discharge sources are much simpler to construct than the others, but much of the beam they produce consists of ground states. Ground-state-to-metastable ratios are typically between $10^5$ and $10^4$ [39, 41, 91, 92]. The thermal or near-thermal velocity beams produced by discharge and electron bombardment (and optical pumping) sources are generally easier to analyze and work with than the fast beams from charge-transfer sources. However, charge-transfer sources produce beams with much lower ground-state backgrounds. Discharge and electron bombardment devices typically require large vacuum pumps because of their high gas loads.

TABLE I. Sources of Metastable Atoms and Molecules

| Atom/ Molecule (Atomic Number) | Primary Metastable States | Types of Sources[a] | Typical Quoted/ Implied Flux/Intensity | References |
|---|---|---|---|---|
| H (1) | $2^2S_{1/2}$ | O, E, C | $10^6$–$10^8$ $s^{-1}$ (O, E); $\sim 10^{11}$ $s^{-1}$ (C) | 22, 24, 26, 38, 49 |
| He (2) | $2^1S_0$, $2^3S_1$ | D, E, C | $10^{13}$–$10^{15}$ $sr^{-1}$ (D, E); $10^{11}$–$10^{13}$ $s^{-1}$ (C) | 7, 9, 11, 12, 18, 29, 30, 39, 43–47, 50–59 |
| N (7) | $2^2D_{3/2, 5/2}$ | D | — | 60 |
| Ne (10) | $3^3P_{0, 2}$ | D, E, C | $10^{13}$–$10^{15}$ $s^{-1}$ $sr^{-1}$ (D, E); $10^{11}$–$10^{13}$ $s^{-1}$ (C) | 8, 14, 16, 29, 39, 41, 42, 48, 51, 54, 61–67 |
| Mg (12) | $3^3P_{0, 1, 2}$ | D, E | $\sim 10^{15}$ $s^{-1}$ $cm^{-2}$ (D) | 68, 69, 70 |
| Al (13) | $3^2P_{3/2}$ | D | $10^{10}$–$10^{12}$ $s^{-1}$ $sr^{-1}$ | 71[b] |
| Ar (18) | $4^3P_{0, 2}$ | D, E, C | $10^{13}$–$10^{15}$ $s^{-1}$ $sr^{-1}$ (D, E); $10^{11}$–$10^{13}$ $s^{-1}$ (C) | 13, 14, 39, 41, 45, 47, 54, 59, 64, 66, 72–74 |
| Ca (20) | $4^3P_{0, 1, 2}$ | D, E | $\sim 10^{15}$ $s^{-1}$ $cm^{-2}$ (D) | 40, 68, 75, 76 |
| Sc (21) | $4^4F_{5/2, 7/2}$ | O | — | 23 |
| Zn (30) | $4^3P_{0, 2}$ | E | — | 69 |
| Ga (31) | $4^2P_{3/2}$ | T | — | 6, 77 |
| Kr (36) | $5^3P_{0, 2}$ | D, E | $\sim 10^{13}$ $s^{-1}$ $sr^{-1}$ (D) | 41, 63 |
| Sr (38) | $5^3P_{0, 2}$ | D | $\sim 10^{15}$ $s^{-1}$ $cm^{-2}$ | 70 |
| Zr (40) | $4^3F_{3, 4}$ | D | $10^{10}$–$10^{12}$ $s^{-1}$ $sr^{-1}$ | 71[b] |
| Nb (41) | $4^5S_2$ | D | $10^{10}$–$10^{12}$ $s^{-1}$ $sr^{-1}$ | 71[b] |
| Mo (42) | $4^5S_2$ | D | $10^{10}$–$10^{12}$ $s^{-1}$ $sr^{-1}$ | 71[b] |
| In (49) | $5^2P_{3/2}$ | T | — | 78 |
| Xe (54) | $6^3P_{0, 2}$ | D | [c] | 73, 79 |
| Ba (56) | $6^3P_{0, 2}$; $5^1D_2$; $5^3D_{1, 2, 3}$ | D | $\sim 10^{15}$ $s^{-1}$ $cm^{-2}$ | 80 |
| Gd (64) | $4^{11}F$, $4^{11}G$ | O | $\sim 10^{15}$ $s^{-1}$ $cm^{-2}$ | 81 |
| W (74) | $5^5D_{0-4}$ | D | $10^{10}$–$10^{12}$ $s^{-1}$ $sr^{-1}$ | 71[b] |
| Hg (80) | $6^3P_{0, 2}$ | D, E | $\sim 10^{12}$ $s^{-1}$ $sr^{-1}$ (D) | 5, 82–84 |
| Tl (81) | $6^2P_{3/2}$ | O, T | — | 85, 86 |
| Pb (82) | $6^3P_{1, 2}$ | D, T | — | 85, 87 |
| Bi (83) | $6^2D_{3/2, 5/2}$ | D | — | 88 |
| $H_2$ | $C^3\Pi_u$ | E | — | 63 |
| $N_2$ | $A^3\Sigma_u^+$ | D, E | $\sim 10^{14}$ $s^{-1}$ $sr^{-1}$ (D) | 47, 54, 63, 89 |
| $O_2$ | $A^1\Delta_g$ | D | — | 63 |
| CO | $a^3\Pi$, $b^3\Sigma^+$ | E | — | 90 |

[a] O—optical pumping; E—electron bombardment; C—charge transfer; D—discharge; T—thermal.

[b] Flux estimates: D. W. Duquette, private communication.

[c] Although references [71] and [78] report no Xe* flux, it can be expected that standard discharge sources would produce Xe* fluxes comparable to those of other noble gases.

Metastable atoms of the metallic or alkaline earth elements are produced most easily in a discharge source, while noble gas metastables are readily made in any source other than one utilizing optical pumping. Hydrogen metastables can be destroyed relatively easily by external fields and are thus usually made in electron bombardment sources. Thermal sources are limited to those elements with low-lying metastable states.

Finally, the velocity profiles of the various source types are very different, and this is often a major consideration when picking a source for a given experiment. Reactions at typical "chemical physics" energies require the lower velocities of discharge or electron beam sources. Between these two, the electron bombardment sources offer better velocity control and resolution than do discharge sources. Charge-transfer sources have by far the best energy resolution, but produce fast metastables that are unsuitable for the study of chemical reactions or the probing of condensed-matter surface phenomena.

Next we discuss in detail the various source types, and then consider techniques for metastable beam modification and characterization. Practical experimental concerns will be addressed throughout, particularly with regard to metastable-associated backgrounds.

## 6.2 Electron-Beam Bombardment Sources

### 6.2.1 Overview

Electron bombardment sources make use of a conceptually simple scheme: the excitation of a beam of atoms to metastable states by a well-defined beam of electrons. Insofar as they effect the metastable intensity and velocity distribution, the important design parameters for such a source are the electron beam energy and current, and its overlap with the atomic beam. Given the rather small cross-section values for metastable excitation ($\lesssim 10^{-17}$ cm$^2$ [93]), intensity considerations demand large electron-beam currents and efficient beam overlap. Unfortunately, the excitation cross-sections are maximal just above threshold, where space charge limitations on the electron beam current density are the most severe. The relative populations of various metastable components within a given atomic beam are also energy-dependent, because triplet excitation falls much more rapidly with electron energy than does singlet excitation. Thus, by 100 eV, virtually all of the metastables produced in, for example, a helium target are in $2^1S$ states.

The electron and atomic beams can be either coaxial or transverse; neither geometry is clearly superior to the other, and other experimental factors not associated with the source often determine which is used. Coaxial sources offer generally larger overlap volumes, but are more complex, typically requiring

more electrodes for electron-beam focusing. Magnetic fields are often used in either configuration to confine the electron trajectories to a well-defined overlap region. Momentum transfer to the atomic beam, in combination with the initial atomic-source velocity profile, is crucial in determining the velocity distribution of the metastables. This issue has been discussed extensively in the literature [44–46, 73, 89]. Early transverse excitation designs suffered from poor velocity resolution; the already broad atomic ground-state distribution characteristic of their effusive sources was further broadened by the momentum transfer perpendicular to the beam resulting from electron bombardment. The first coaxial sources improved velocity resolutions somewhat. Recent sources of both transverse and coaxial design, using supersonic atom beams, geometric or mechanical velocity selection, pulsing of the atomic and/or electron beams, or a combination of these, have succeeded in producing $\Delta v/v$ values approaching 3%. Velocity spreading due to electron bombardment is less when heavy species such as Ar are used instead of H or He. Mean velocities can be modified by using resistive heating or conductive cooling [11, 46, 74] of the atomic beam source. Indeed, one of the major advantages of electron bombardment sources is that velocity distributions can be controlled with relative ease.

### 6.2.2 Coaxial Geometry

A state-of-the-art coaxial electron-beam source, used for producing metastable noble gas beams, is shown schematically in Figure 1 [45]. It uses a supersonic nozzle atomic beam source, downstream (~15 mm) from which is a 0.6 mm diameter skimmer. The atomic stagnation temperature and pressure are nominally 300 K and several atmospheres, respectively. To avoid overloading the vacuum pumps, the atomic source is pulsed using an automobile fuel injector, with typical pulse-time widths of tens of milliseconds.

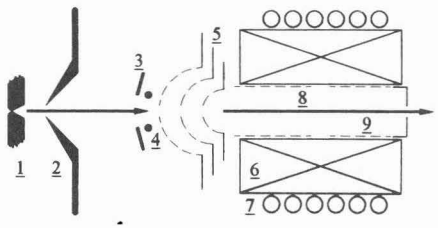


FIG. 1. Schematic diagram of the coaxial electron-beam bombardment metastable source of Kohlhase and Kita [45]. Shown are (1) pulsed supersonic nozzle ground-state atom source; (2) skimmer; (3) electron repeller; (4) electron filament; (5) extractor, control, and acceleration grids; (6) electromagnet; (7) cooling water coil; (8) first anode; (9) second anode.

The atomic beam enters an excitation volume whose first element prevents electrons from migrating upstream. Downstream, a thermionic cathode is looped to just circumscribe the atomic beam, and three hemispherical mesh electrodes accelerate and pulse the electron beam. The main collision volume, surrounded by a water-cooled solenoid, has a two-part mesh anode and an exit aperture. The electron beam pulsing is used in place of a mechanical chopper to allow metastable time-of-flight (TOF) information to be obtained.

The natural velocity percentage width of supersonic nozzle sources is generally less than 10%, and the coaxial excitation does not affect this distribution significantly. This source is reported to have a velocity width of between 3 and 4% for He, comparable to the best results reported to date. It is also reported to produce an extremely high instantaneous metastable flux: with Ar the authors quote a value of $5 \times 10^{15}$ atoms $s^{-1}$ $sr^{-1}$. It should be noted that coaxial sources can suffer velocity broadening due to production of fast metastable negative ions which are neutralized in collisions with ground state atoms before exiting the source. This process yields a second velocity component of the output beam [72].

### 6.2.3 Transverse Geometry

Tommasi *et al.* [44] have described a metastable source with transverse excitation geometry. In their apparatus, shown schematically in Figure 2, the momentum transfer due to electron-impact excitation is turned to an advantage by using it to substantially separate the supersonic ground-state beam from the metastable component produced in the electron gun.

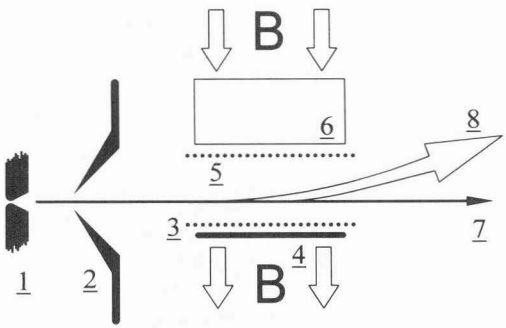


FIG. 2. Schematic diagram of the transverse electron-beam bombardment metastable source of Tommasi *et al.* [44]. Shown are (1) pulsed supersonic nozzle ground-state atom source; (2) skimmer; (3) electron control grid; (4) indirectly heated planar cathode; (5) electron control grid; (6) parallel-plate slotted anode; (7) undeflected ground-state beam; (8) collisionally deflected metastable atoms. The electron bombardment volume is immersed in a uniform magnetic field.

A piezoelectric valve produces pulses with a repetition rate of ~2 Hz and a width of <1 ms, corresponding to $\sim 10^{18}$ atoms/pulse. The duty cycle is again limited by pumping speed. After collimation by a skimmer, the atom beam enters a region where it is crossed by a 5 mm × 25 mm electron beam, defined by the emitting area of the cathode. The cathode is specially treated with barium–strontium carbonate, using a recipe developed to provide a very robust emitting surface [46, 47]. Electron energies are controlled by two grids and are kept near the metastable excitation threshold to minimize collisionally induced spatial spread of the beam. The electron beam is dumped into an anode comprising an array of parallel plates, designed to minimize the effects of space charge. This design permits electron currents greater than 0.1 A to be produced, even though the intergrid gap of 11 mm, dictated by the necessity of preventing metastable–grid collisions, is rather large.

The He or Ar metastable beam thus produced is reported to have extremely high instantaneous flux ($>10^{15}$ s$^{-1}$ sr$^{-1}$) and has a FWHM divergence of less than 5°. The percentage velocity width of the beam is 7%, due almost entirely to the ground-state source velocity spread. Moreover, the deflected beam population is actually inverted, i.e., over a restricted angular range, the flux of metastables exceeds that of ground state atoms. In addition, Tommasi *et al.* have observed spatial separation within the He* beam of the $2^1S$ and $2^3S$ states. This is due to the different excitation thresholds of the two species, which cause kinematic shifts in their trajectories. The $^3P_2$ and $^3P_0$ states of the heavy noble gases would exhibit a similar separation, although it would be smaller than in He because of the correspondingly smaller energy splittings.

## 6.3 Discharge Sources

### 6.3.1 Overview

A large variety of discharge sources have been developed, based on a broad range of geometries and discharge types. While low-current ($\lesssim$1 A) cold-cathode sources are the most commonly used, other types, involving pulsed, RF, and even audio frequency discharges have been reported. Often they are outgrowths of molecular dissociators or arc-heated fast atom sources, in which metastable fractions are considered a nuisance. While discharge sources are generally simpler to build and operate than electron bombardment devices, they suffer from broader velocity distributions which are less easily controlled and which are dependent on discharge polarity, current, and voltage. Another potential problem is the large ground-state fraction present in beams from discharge sources; the metastable-to-ground-state ratio is typically less than $10^{-4}$. Discharge-induced sputter erosion is the limiting factor in source lifetime, as opposed to gun filament life in electron beam sources. In atomic scattering experiments,

photon backgrounds from the discharge can be particularly severe. Sources of this type can be classified very roughly as belonging to one of the following categories.

### 6.3.2 Hot-Filament-Cathode Effusive Sources

These are used for gaseous samples. The metastable atoms effuse from an aperture in the plate anode opposite the cathode (Figure 3a). Fluxes are low ($\lesssim 10^{10}$ metastables $s^{-1}\,sr^{-1}$), with velocities determined by the discharge temperature, which is determined in turn by power input to the cathode. Fast atom components can result from collisional neutralization of metastable negative ions which have been accelerated toward the anode.

### 6.3.3 Effusive Hollow-Cathode Discharge/Sputter Sources

In these sources (Figures 3b–3d), a DC discharge is struck between a hollow cathode and an anode which may or may not define the output beam. Two disparate examples of this type are that of Theuws *et al.* [41], in which the cathode acts as the tube from which the parent gas effuses, and that of Duquette and Lawler [71], which has been used to produce metastable metal atoms. In the latter source, argon is used to maintain a discharge between anode and cathode, and argon ions sputter a metal sample which coats the cathode. The free metal atoms are subsequently excited in the discharge, and some effuse from an exit aperture in the anode. A variant on this type of source, used to produce alkali-earth metastables, has been reported by Ureña *et al.* [40]. Ca is heated in a cylindrical crucible to ~1400 K and effuses into a discharge region between the crucible and the concentric, electrically isolated cylindrical oven. There it is excited and emerges from the oven. The discharge can be pulsed if desired. Large Ca* fluxes have been reported for this source. A similar source with an axial beam geometry has been reported by Brinkmann *et al.* [75].

### 6.3.4 Gas-Dynamic Sources

A unique source, developed by Brinkmann and co-workers [76], was also used for Ca (Figure 3e). The Ca is vaporized in a cylindrical oven and diffuses into a hollow stainless-steel cathode through holes drilled in the cathode wall. The discharge runs between the cathode and an anode made of wire wrapped in a spiral whose axis is coincident with that of the cathode. Along this axis is directed a "carrier" beam of argon, which is passed through a filament preheater prior to entering the hollow cathode. The metastable atoms are thus carried out of the discharge and attain a velocity distribution characteristic of the argon,

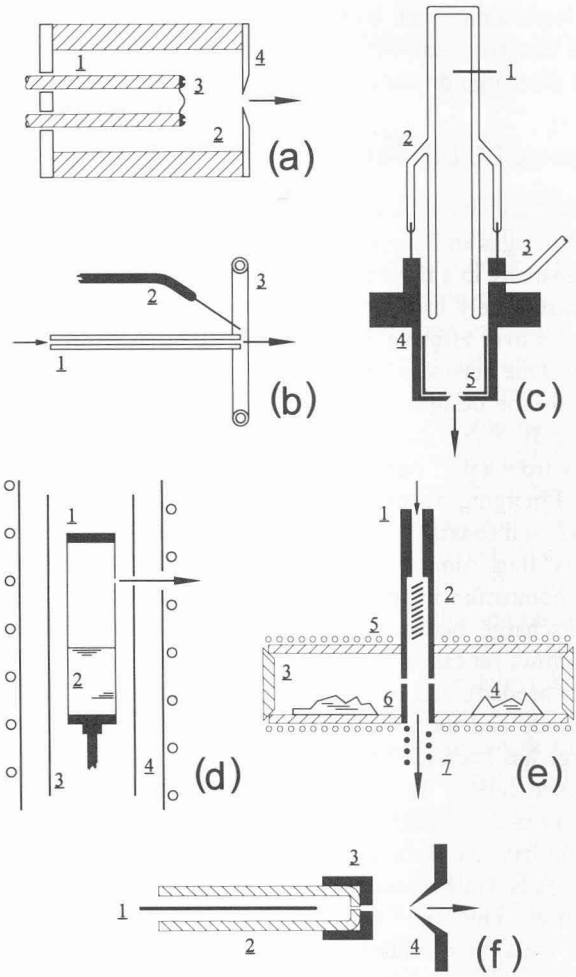


FIG. 3. Various configurations of discharge metastable sources. (a) Hot-cathode effusive source [7, 8] showing (1) insulating cathode supports; (2) insulating discharge container; (3) hot cathode; (4) anode. (b) Hollow-cathode arc source [41] showing (1) hollow cathode; (2) water-cooled ring anode; (3) ignition electrode. (c) Hollow-cathode sputter source [60] showing (1) anode; (2) argon column; (3) argon inlet; (4) hollow cathode; (5) sputter target. (d) Effusive oven source [40] showing (1) crucible (anode); (2) metal to be vaporized; (3) hollow cathode; (4) water-cooled heat shield. (e) Gas-dynamic jet source [76] showing (1) argon inlet; (2) preheater; (3) oven; (4) sample to be vaporized; (5) oven heater coils; (6) hollow cathode with effusive entrance apertures; (7) coil anode. (f) Supersonic cold cathode discharge source of Brand *et al.*, incorporating improvements on the design of reference [64] showing (1) W cathode rod; (2) alumina tube with laser-drilled nozzle; (3) aluminium cap anode; (4) skimmer and secondary anode.

which can be supersonic when an appropriate nozzle is used. Electrons carried away from the discharge can be used to seed a second discharge downstream. The additional discharge dramatically increases the metastable production [43].

### 6.3.5 Supersonic DC Discharge Sources

These sources, which are among the simplest to build and operate, are based on the original designs of Searcy [52] and Leasure *et al.* [53]. They have been developed extensively in a number of laboratories [9, 25, 28, 29, 39, 42, 50, 51] and are the most widely used source type today. An advantage of supersonic discharge sources over effusive ones is the relatively narrow velocity spread of the metastables. One version of this source type, representing modifications and improvements on the designs of Fahey *et al.* [54] and of Brand *et al.* [61], is shown in Figure 3f. Noble gas with a stagnation pressure of 1–2 kPa emerges in supersonic flow from a 0.15 mm diameter nozzle that has been laser-drilled in an alumina tube. Emerging atoms are excited by a discharge struck between a tungsten cathode rod coaxial with the alumina tube and an aluminum-cap anode. This source is thus similar in concept and excitation geometry to the electron-beam bombardment source of Kohlhase and Kita [45]. Earlier designs relied on a discharge between the skimmer and the cathode. The present configuration allows for easier initiation of the discharge (no high-voltage pulser or Tesla coil is needed), and a second discharge can, if desired, be maintained between the anode and the skimmer, which is electrically isolated. A second in-line discharge has been used both in the hot arc source of Ferkel *et al.* [43] and, as mentioned earlier, the gas dynamic source of Brinkmann *et al.* [76] to significantly increase metastable production. Liquid nitrogen cooling of the discharge region has also been used effectively to reduce metastable velocities and velocity spreads while increasing flux [20, 42]. When the source's polarity is reversed, positive ions are accelerated towards the skimmer, and fast ground-state or metastable neutrals can populate the beam [52, 72, 91].

Various authors have reported the metastable intensity and fine-structure distribution (e.g., the ratio of $2^3S_1$ to $2^1S_0$ populations in He) to depend on stagnation pressure and discharge current and voltage [28, 39, 41–43, 51, 61, 72, 95]. Some designs use coaxial or radial magnetic fields to enhance the discharge and hence metastable intensity. For this type of source, however, the two most crucial factors appear to be the vacuum in the region between the nozzle and skimmer, and the nozzle/skimmer alignment [39, 61]. The vacuum is important because elastic collisions with background gas remove metastables from the beam. Thus, skimmer geometry and nozzle/skimmer distance can also be important, because any supersonic beam reflected from the upstream skimmer walls will act as residual background gas. The source in Figure 3f has a nozzle operated in a confined chamber ($<1$ liter volume) pumped by a 300 liter/s

turbomolecular pump. The aluminum cylinder is mounted on a robust precision $x$–$y$ translational stage for alignment. These design features have been instrumental in obtaining high metastable fluxes.

## 6.4 Charge-Transfer Sources

A serious problem that can occur with electron-beam and discharge sources is background due to the large ground-state component accompanying the metastable fraction. For example, in electron-metastable scattering experiments, such a ground-state component means that incident electron energies must be kept below (depending on the specific experiment) the first excitation threshold for optical decay or the ionization threshold of the ground-state atoms. In the case of He targets, this limits incident electron energies to the range below 25 eV. Charge-transfer sources produce ground-state-to-metastable ratios closer to $10^0$ than to $10^5$, and thus eliminate a large part of this problem [49, 59, 65, 66].

Virtually all of the charge-transfer sources reported have been used for the production of hydrogen or noble gas metastable atoms, with the alkali metals, parent noble gases, or $H_2$ used as the charge-transfer target. The charge-transfer and metastable production process proceeds most efficiently when two conditions are met: the energy defect, $\Delta E$, in the production process is as small as possible, and the incident ion velocity is such that electron velocity matching between the initial and final states occurs [36, 59]. The defect $\Delta E$ is defined as the difference in ionization energy between the target ground state and the projectile metastable state. Velocity matching simply means that the electron to be transferred need not undergo large acceleration during the collision.

The velocities required for the best metastable production thus imply the use of high-voltage accelerators, which, in conjunction with the use of ion sources, means that efficient charge-transfer sources are relatively complicated. Moreover, the fast metastables and the accompanying ground-state component can excite background gas in the experimental interaction region, leading to another source of background. Also, the fast beam is more difficult to manipulate with lasers (Section 6.7).

A recently reported state-of-the-art source, which has produced very high Ne* fluxes, is shown schematically in Figure 4 [36, 67]. $Ne^+$ is produced in a standard water-cooled Penning ion source, which is floated at $+800$ V with respect to ground. The extracted, space-charge-limited beam of about 35 μA is accelerated and periodically focused by a series of Einzel lenses which transport the beam to the charge-transfer cell and serve to decouple the rest of the experiment from the effusive gas load of the ion source. The ion beam traverses a Na vapor target 1 cm thick at a nominal pressure and temperature of 0.03 Pa and 500 K, respectively. The neutral flux leaving the charge exchange cell is $\sim 10^6\ s^{-1}\ sr^{-1}$, of which 50% are in a metastable state. The metastable density

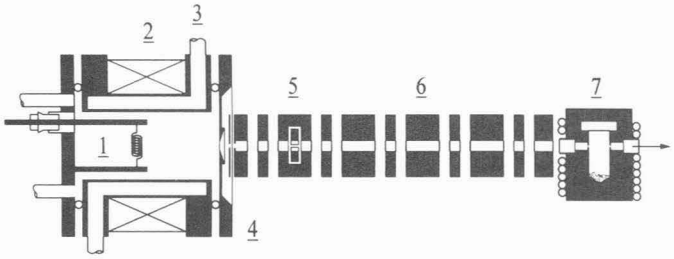


FIG. 4. Schematic diagram of high-flux charge-transfer metastable source [36, 95] showing Penning ion source floated at +800 V with respect to ground: (1) hot anode; (2) electromagnet; (3) cooling water. Also shown are (4) extraction cathode; (5) electrostatic deflection electrodes; (6) transport Einzel lenses; (7) grounded charge–transfer cell.

at the interaction region 25 cm downstream from the charge exchange cell is $\sim 10^6$ cm$^{-3}$. If Ne is used in the charge exchange cell instead of sodium vapor, the source can produce a fast ground-state beam. Source lifetime is limited by Na coating of Einzel lens element insulators, which causes electrical breakdown after several hundred hours of operation.

An interesting variation on the standard charge-transfer vapor target is the use of a graphite multichannel "converter," developed by Gostev *et al.* [57, 58]. They describe a device in which $He^+$ from a Penning source is neutralized with 25% efficiency into the $2^1S$ and $2^3S$ metastable states. The graphite array also serves to collimate the atomic beam and control its velocity by acting as the extraction electrode for the ion source. This apparatus was reported to produce extremely high steady-state fluxes of He* ($\sim 10^{20}$ s$^{-1}$ sr$^{-1}$), although more recent analyses have cast doubt on this number [36]. Moreover, attempts to use it to produce metastable species of the heavier noble gases led to very short source lifetimes due to sputtering of the converter material. Discussions of other charge-transfer sources are contained in references [13–19, 24, 48, 49, 59, 65, 66].

## 6.5 Optical-Pumping Sources

Metastable atoms can be produced in two-step "optical pumping" processes involving excitation of the ground state to an excited state by photon impact, followed either by collisional "quenching" or photon decay to a metastable level. The latter process has the advantage that neither the excitation nor the decay processes involve significant momentum transfer, and the resultant metastable velocity profiles can thus be very narrow. Optical production of metastables has the general advantage over bombardment and discharge sources that by tuning or filtering the photon source, specific metastable species can be produced. (This is particularly simple if dye lasers are used.) Sources using optical pumping have been reported for Hg [5, 82], Sc [23], Tl [86], H [38], and Gd [81]. With the

exception of the Sc and Gd sources, which used dye lasers, discharges containing the parent atom were used to provide the pumping photons.

In the Hg source of Haberman *et al.* [82], Hg vapor produced in an oven is combined with nitrogen and the mixture is passed coaxially at a pressure of ~10 kPa over a cylindrical Hg resonance lamp. The 254 nm radiation from the lamp excites Hg $6^3P_1$ states. Some of these are subsequently converted to metastable $6^3P_0$ states through collisions with $N_2$ molecules. At the end of the lamp, the Hg-seeded nitrogen is formed into a beam by passing through a nozzle and skimmer. In addition to its role in collisional production of the $6^3P_0$ metastables, the $N_2$ also serves to accelerate and narrow the velocity distribution of the Hg as it passes through the nozzle. The $N_2$ must be kept very pure to prevent $O_2$ and hydrocarbon quenching of the Hg metastables. With this source, Hg* fluxes of $4 \times 10^{12}$ s$^{-1}$ sr$^{-1}$ were observed.

Beams of metastable hydrogen atoms have been produced in a similar fashion, but with somewhat more difficulty, given that atomic hydrogen must first be produced from $H_2$ (e.g., by RF discharge) and formed into a beam [38]. Lyman $\beta$ radiation (103 nm) from a second RF discharge is used to pump the beam to the $3P$ state, with subsequent decay to the metastable $2^2S_{1/2}$ level. Particular care must be used in the case of hydrogen to avoid external stray fields that can quench the metastable levels by motional or static Stark mixing with the $2P$ state. The use of windows for Ly$\beta$ radiation and of Ly$\alpha$ detectors to monitor the metastable production complicate matters further. Harvey [38] reports production of a metastable H beam with intensity $10^6$ s$^{-1}$ using this technique. This value is comparable to that typical for electron-bombardment sources.

## 6.6 Thermal Sources

Many atoms have ground states which are the lowest components of fine-structure multiplets. If other components of this multiplet are metastable and are not too far above the ground-state in energy, the effusive beam from an oven source can have a significant metastable fraction. Hishinuma and Sueoka [6], for example, describe a graphite oven source for Ga ($4^2P_{3/2}$) metastable effusive beams which operates at 1600 K. The splitting between the ground $4^2P_{1/2}$ and $4^2P_{3/2}$ metastable states in Ga is $\Delta E = 0.10$ eV. Thus, a Boltzmann distribution with appropriate statistical weights yields a ratio for metastable-to-ground-state atoms as

$$\frac{N^*}{N_g} = \frac{g^*}{g_g} \exp\left( + \frac{\Delta E}{kT} \right) = 0.47, \tag{6.1}$$

where $g^*$ and $g_g$, the statistical weighting factors, are 4 and 2 for the metastable and ground states, respectively. Geesmann *et al.* [85] and Bartsch *et al.* [78] have reported observing metastables in Th and Pb from a thermal source operated

between 1000 K and 1400 K. Other elements which have the appropriate multiplet structure for such sources include, B, Al, In, Cl, Br, I, and Lu.

## 6.7 Beam Modification and Analysis

Beams of particles emerging from metastable sources generally contain ions, fast or slow neutrals in a variety of excited states, ground-state neutrals, and electrons, as well as the metastable species. In addition, more than one metastable state may be produced, as is the case with noble gas beams. It is thus important to characterize and possibly alter the emergent beam. This can be done in a variety of ways, which depend primarily on the metastable atom or molecule of interest. A diagram illustrating a generic gauntlet of diagnostic devices is shown in Figure 5. We note that fast metastable beams, such as those produced by charge transfer, are more difficult to alter by means of photons or inhomogeneous (Stern–Gerlach or six-pole) magnetic fields than are thermal beams [48, 96].

### 6.7.1 Collimation and Charged Particle Removal

Discharge and thermal sources require some collimation immediately following the beam production region. This is typically accomplished with a skimmer, whose geometric profile can be quite important [39], and which often serves as one of the discharge electrodes. In addition to reducing the divergence of the beam, collimators also substantially reduce background photons. Electron-beam, optical, and charge-transfer sources more often rely on collimation of the parent beam prior to excitation. Electrons and ions can then be removed from the beam by electric field plates or a charged grid [18]. This technique has the ancillary benefit that atoms in highly excited Rydberg levels, which might not decay to a ground or metastable level over the normal flight path of the apparatus, are field-ionized [97]. A simple drift region after the source can also be effective in removing many of the shorter-lived excited states.

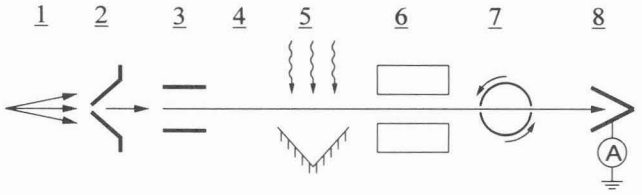


FIG. 5. Schematic gauntlet of beam modification and diagnostic devices (see text): (1) divergent metastable beam from source; (2) skimmer; (3) field quenching/ion deflection plates; (4) drift region; (5) optical pumping/quenching region; (6) inhomogeneous magnetic field for state selection or analysis; (7) velocity selector/analyzer; (8) metastable detector.

### 6.7.2 Metastable Quenching and State Selection

If more than one metastable species is produced by the source, it is often desirable to "quench" all but one of these states. This is accomplished most readily by photon excitation of the levels to be eliminated. The excited states can decay subsequently either directly or indirectly to the ground state. Helium $2^1S$ states are usually quenched using 2.06 μm light from a He discharge tube wrapped in a spiral about the beam [9, 30, 43, 46, 98]. This light induces transitions to the $2^1P$ state, which can decay to the ground state. A high-power Ti-sapphire laser could also be used for this purpose. The $2^3S$ states cannot be destroyed except by collision processes. Tommasi *et al.* [44] use the different momentum transferred to the He atom upon electron impact creation of $2^1S$ and $2^3S$ states to separate the two metastable components spatially. Lasers have been used to eliminate either the $^3P_2$ or $^3P_0$ metastables in heavy noble gas beams [48, 51, 61, 99].

Once a single metastable variety has been isolated, further state selection can be accomplished either by additional optical pumping or by manipulation with inhomogeneous magnetic fields. A simple Stern–Gerlach magnet can be used to select and/or analyze metastables with a given magnetic quantum number, albeit with significantly reduced intensity [7, 8, 28, 30]. Six-pole magnets have been used very effectively to produce highly polarized He ($2^3S$) beams [25]. Baum *et al.* [28] used a supersonic DC discharge source of He* succeeded immediately by a skimmer and six-pole magnet with a central "stop," or plug placed on the beam axis. Ground-state atoms (which make up the vast majority of the emergent beam) and $2^1S$ states, both of which have $M_J = 0$, are undeflected in the magnet and are largely stopped by the axial plug. He ($2^3S$) states are not only polarized by the six-pole [96], but are also focused past the stop, so that their intensity on-axis is enhanced. Baum *et al.* report metastable polarizations of 90%, with ground-state-to-metastable ratios as low as $10^2$. Six-poles are generally ineffective with fast beams from charge transfer sources.

Optical pumping can also be used to polarize the metastables [100]. (See also Chapter 9.) Consider as an example the $^3P_2$ states of the heavy noble gases. If circularly polarized radiation is used to drive multiple transitions between the $np^5(n+1)s\ ^3P_2$ and $np^5(n+1)p\ ^3D_3$ levels, the $^3P_2$ state will become oriented in the direction of the incident photon angular momentum. Once a beam has been spin-polarized, the extent of its polarization can be measured using, e.g., a Stern–Gerlach magnet.

### 6.7.3 Beam Compression

In addition to their utility for state selection, photon beams and six-pole magnets can also be used for beam compression. As mentioned in Section 6.7.2, the use of a six-pole magnet for this purpose has been demonstrated [28].

Recently, laser cooling and compression of noble gas metastable beams has been demonstrated [20, 21]. Detailed discussions of such techniques are presented in these references and in Chapter 8 of this volume.

### 6.7.4 Velocity Selection and Analysis

An emergent beam's velocity profile can be analyzed in a variety of ways. For a DC source, a mechanical slotted chopper in conjunction with time-of-flight (TOF) analysis is often used [9, 43, 51, 54, 72]. This method is useful for establishing the beam fraction due to photons and fast neutrals. For pulsed beams, TOF techniques are also applicable [40, 45, 64]. A number of authors have reported using laser absorption measurements to determine velocity distributions [42]. By monitoring the light absorption as a function of photon incidence angle, and hence Doppler shift, the metastable velocity distribution can be mapped.

Velocity selection can be done completely mechanically or with a combination of pulsing and mechanical chopping with good resolution, but with some intensity diminution [6–8, 101]. The pulsed rotor source of Simons *et al.* [73] provides the simplest way to accomplish crude mechanical velocity selection. Generally, velocity selection is best accomplished by adjustment of the velocity profile of the initial (parent) ground state atoms, prior to excitation [11, 20, 46]. Such velocity control is sufficient for most applications.

### 6.7.5 Intensity Measurements

The topic of intensity and flux measurement is taken up on detail in Chapter 11. We simply note here that a variety of techniques can be used, including chemical reactions [3, 66, 98], laser-induced fluorescence and quenching [36, 42, 102], static electromagnetic field quenching [22, 38], and Auger electron emission from solid surfaces [31, 36, 82, 103–106]. These disparate techniques must be carefully calibrated if two of them are used to compare source intensities. Specifically, absolute fluxes are difficult to measure using the Auger method because of the uncertainties in secondary electron emission coefficients and the wide range of assumptions made in using them to estimate source strength.

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