New methods in molecular beam studies

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A brief review is given of the development of methods used in investigations employing molecular beams. The topics discussed include the latest achievements in the development of gas-dynamic sources of molecular beams, the use of lasers in investigations employing beams, new methods of time-of-flight analysis of energies of particles in beams, and new possibilities associated with the use of position sensitive detectors based on secondary emission and utilizing microchannel plates.

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Studies of atomic and molecular processes based on the scattering of beams of atomic particles have yielded an enormous amount of new data during the last few years. The range of applicability of such data is exceedingly extensive and runs from interpretations of atomic and molecular processes in the interstellar medium and in the upper planetary atmospheres to problems involving the optimization of the most recent power sources such as powerful gas lasers, MHD generators, and so on.

Although the available quantitative results have been examined and generalized in a large number of review papers (see, for example, Refs. 1 and 2), methodological aspects of such studies have, as a rule, been undeservedly ignored despite the fact that they are of intrinsic value since, in the final analysis, a new method is practically always a prerequisite for a new result. A thorough knowledge and utilization of new methods is one of the main ways of increasing the effectiveness of any research laboratory.

In view of the foregoing, our aim in this review will be to examine the "technological aspects" of scattering experiments that are currently being performed in atomic and molecular physics.

It is important to note that the techniques used in studies of atomic and molecular scattering have undergone both qualitative and quantitative changes in recent years. Even relatively recently, i.e., at the time of the publication of Ramsey's well-known book, "Molecular Beams," the size of the equipment used only occasionally exceeded 1 m in one dimension. The apparatus now used in much larger because, on the one hand, the vacuum conditions are now much better, especially in the region of the beam detector, and, on the other hand, the diagnostic equipment is now more varied and more complicated. The crossed-beam installation developed at the Institute for Aerodynamic Studies at Goettingen, West Germany, is an illustration of the scale and complexity of modern experiments. The integral diameter of the vacuum chamber is 1.4 m and its height is 4.3 m. There are four rotatable flanges (one has a diameter of 1.4 m) used to vary the angle between the two beams and the angles at which the collision products are observed without releasing the ultrahigh vacuum.

A complete review of new methods is well-nigh impossible and we shall therefore confine our attention to the following topics:

1a) new achievements in the gas-dynamic method of beam production,

2b) the use of lasers in studies involving molecular and atomic beams,

3c) new techniques in time-of-flight analysis of the energy spectra of beams, and

4d) new detection techniques based on microchannel plates.

1. The gas-dynamic method of beam formation is based on the phenonemon of free expansion of a gas jet as it enters a vacuum. A special conical diaphragm can be used to let out only the core of the jet which is thus transformed into a beam. The method was first proposed and realized already in 1951,[3,4] and has been examined in detail by Leonas.[5]

Since then, the method has been improved in important and striking ways, illustrating particularly clearly the universal value of developing new techniques.

In fact, whilst the data listed in Table I and shown in Fig. 1 illustrate some of the possibilities of laboratory applications of this method, its technological importance is indicated by the report that an experimental plant has been built in South America for the separation of uranium isotopes, using the outflow of a jet into a vacuum.

Figure 1 summarizes the parameters of an H_2 beam under different outflow conditions characterized by the product p_0d_0 (p_0 is the pressure head and d_0 is the diameter of the nozzle) and exhibits the measured time-of-flight spectrum of particles in an He beam.[6]

It is clear from the nature of the spectrum and from the data in Table I that the "natural width" of the velocity spectrum for large values of p_0d_0 is comparable with and, perhaps, even better than that achieved by a mechanical selector. This means that there is no need to use such selectors, and an additional gain can be achieved in the intensity of the monokinetic beam.

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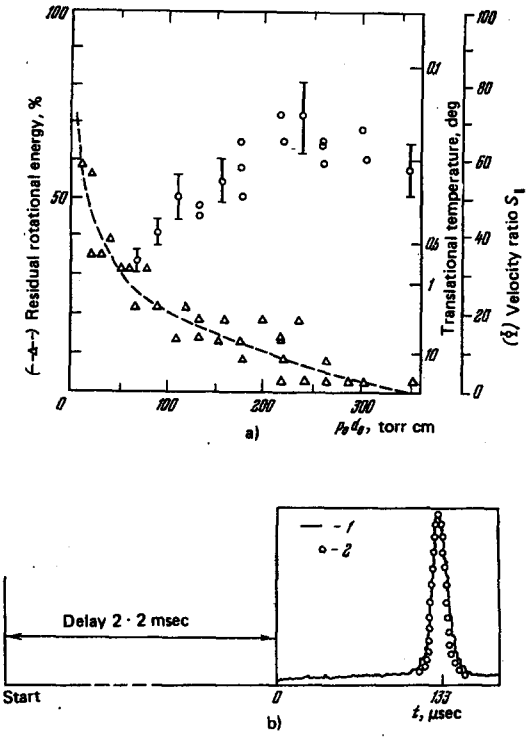

FIG. 1. a) Properties of an H_2 molecular beam produced by a gas-dynamic source (nozzle diameter $d_0 = 25.5\ \mu$, diameter of conical diaphragm $d_c = 0.5$ mm, temperature of initial gas $T_0 = 293$°K, $S_{\parallel}$—ratio of velocities of directed and random motion in the beam[6]); b) time-of-flight spectrum obtained for an He beam with the same source [$T_0 = 320$°K, $p_0 = 200$ bar, 1—measurement, half-width at half-height = 12.5 μsec; 2—calculation with $S_{\parallel} = 340$, $T_{\parallel} = 6.5 \times 10^{-3}$ °K (Ref. 6)].

Table I summarizes some of the best characteristics of gas-dynamic sources achieved with a heated working gas and using the well-known effect of acceleration of the heavy component[5] when the emerging jet consists of a light gas (carrier) and an admixture with a higher molecular weight.

The gas-dynamic method has been effectively employed to produce beams not only of stable but also of chemically unstable atoms (Cl[6], Br[7], H[8], N[9], O[10], and Cu[11]). Such beams are usually accompanied by other components (carrier gas and parent molecules), but selective detection can be achieved with the aid of a quadrupole mass spectrometer which is now a standard instrument in laboratories employing molecular beams.

The above data are not an exhaustive summary of all the possibilities of the gas-dynamic method. If molecules M (for example, M = NO_2, I_2, CsF) are added to the outflowing gas X, it is possible to achieve both acceleration and cooling of the rotational degrees of freedom of the added molecules (vibrational levels are not, as a rule, excited in the initial state).

TABLE I. Limiting parameters of beams produced by the gas-dynamic method.[6]

Intensity	
Ar beam	10^{19}—10^{20} atom sterad^{-1} sec^{-1}
He, He_2 beam	10^{20}—10^{21} atom sterad^{-1} sec^{-1}
Energy range of beam particles	10^{-2}—40 eV
Beam velocity spread ($\Delta v/v$, He)	0.5%
Corresponding Mach number (S_1)	340
Equivalent temperature $T_{\parallel}$	$6.5 \cdot 10^{-3}$ °K

Since rotational degrees of freedom relax quite readily (Fig. 1a), the cooling of the expanding jet will practically completely "freezout" any initial rotational excitation of the molecules. Moreover, triple collisions (M–M–X) which are dangerous from the point of view of condensation are suppressed, and one can readily avoid condensation of the molecular gas supercooled to 1–10 °K (the boiling point of NO_2, is 300 °K). When a beam of this kind is used in molecular absorption spectroscopy, we have the unique possibility of being able to investigate lines due to free molecules instead of the molecular bands that are difficult to resolve. This possibility has been reviewed by Kitaev and Mal'tsev.[12]

Such "cold molecules" can be observed in their free state only in the interstellar medium (clouds). This medium is now "reproduced" in the laboratory and it is difficult to overestimate the importance of this for the radio astronomy of molecular lines and the submillimeter astronomy which is beginning to emerge. This new technique is also potentially important for studies connected with the problem of laser separation of isotropic molecular compounds of uranium.

We note that the intensity of the fluorescence emitted by the molecular jet under real conditions[13] is such that, even when the Fabry-Perot interferometer is used for its spectral analysis, the discrete photon-counting rate is 10^5–10^8 sec^{-1}.

We conclude our review of the unique possibilities associated with the free expansion of a jet into a vacuum by recalling the phenomenon of collisional polarization of molecules in a gas-dynamic source.[14]

Since intermolecular forces are anisotropic, the scattering cross section is a function of the orientation of the molecule with respect to the relative velocity of the two partners in collision. Even a simple qualitative analysis of the connection between the orientation of a linear molecule and the angular momentum vector J suggests the possibility of an orienting-polarizing effect in the case of nonequilibrium outflow into a vacuum. The origin of the polarizing effect can be more readily understood if, following Ref. 14, we consider the scattering of a beam of molecules with velocities $\mathbf{v}_1$ over a length dx as a result of collisions with particles having velocities $\mathbf{v}_2$ and density n_2. The reduction in the intensity I of the beam is then given by the usual relation

$$dI = -\frac{I g n_2 \sigma\, dx}{v_1},$$

where $\mathbf{g} = \mathbf{v}_1 - \mathbf{v}_2$ is the relative velocity, $\sigma = \sigma_0[1 + \beta P_2(\mathbf{gJ})]$ is the total scattering cross section in the case of isotropic interaction, β is the anisotropy parameter, and P_2 is the second-order Legendre polynomial. The total reduction is obtained by averaging over the relative velocity distribution:

$$\Delta I = -\frac{I \sigma_0 n_2\, dx}{v_1} \langle [1 + \beta P_2(\mathbf{gJ})]\, g \rangle.$$

It is now clear that the additional quantity βP_2 will

be averaged out in the case of isotropic distribution of relative velocities and, hence, the polarizing effect will vanish also. However, when the relative velocity distribution is anisotropic, and this is characteristic precisely for the outflow of a mixture of gases with different molecular weights, the anisotropic contribution will not be lost through averaging, and there will therefore be an appreciable polarizing effect.

The most attractive feature of this polarization effect is that it can be used for symmetric molecules with zero electric dipole moment.

2. Investigations involving the use of laser beams began in recent years and are rapidly developing. The implications of the availability of tunable lasers are only just emerging, but even the initial results are very impressive.

It is possible to define four different ways of using laser beams in molecular beam studies:

a) time-of-flight measurements based on the observation of laser-induced fluorescence,

b) preparation of targets with given internal state,

c) ultrahigh-resolution molecular spectroscopy, and

d) analysis of dynamic and energy states of collision products through selective laser detection.
collision products through selective laser detection.

All these applications are based on the phenomenon of fluorescence induced by highly monochromatic tunable lasers. The resonance nature of the absorption process ensures high selectivity both with respect to the chemical identity and the energy states of the detected particles.

The simplest arrangement for time-of-flight measurements (Fig. 2a) involves the usual chopping of the molecular beam at the beginning of the drift space and

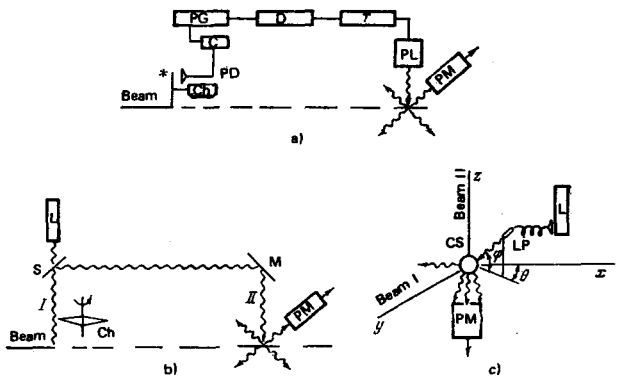

FIG. 2. Schematic illustration of laser fluorescent diagnostics of beams: a) time-of-flight analysis by the excitation method (Ch–chopper, PD–photodiode, C–counters, PG–pulse generator, D–delay line, T–trigger, PL–pulsed laser, PM–photomultiplier); b) time-of-flight analysis using pumping and subsequent excitation (S–laser beam splitter, L–laser, M–mirror); c) Doppler spectroscopy method for the simultaneous analysis of velocities, chemical compositions, and energy state of collision products (CS–center of scattering, LP–light pipe).

the observation of the time evolution of the fluorescence signal at the final point where the molecular and the laser beams intersect. Ion collection in the usual ionization detector is now replaced by the collection of the secondary fluorescence photons.

One of the advantages of this method of detection is that the demands imposed on the background pressure in the region of detection are less stringent, which cannot be ignored in view of the difficulty of maintaining the necessary vacuum in the region of the detector.

A more elegant (and no less universal) method is that used by Gaily *et al.*[15] and illustrated schematically in Fig. 2b. Here, the probing laser beam is divided into two beams, one of which (beam *I*) illuminates the beam atoms at the initial point and the other (beam *II*) at the final point of the drift space.

Beam *I* is modulated by a rotating paddle and is used to pump up the beam particles, for example, Na_2, into an excited electronic state ($B^1\Pi_u, v = 6, j = 43$) for which the probability of relaxation to the initial vibrational-rotational levels of the ground state $X'\Sigma_g^+$ is small. Since there are no collisions in the beam, the illuminated particles retain their state (which is different from the initial vibrational-rotational state) at the end of the flight path, so that the fluorescence signal in beam *II* is exclusively due to particles shielded by the paddle and not subjected to the optical pumping.

The use of Doppler shift provides us with another method of varying the particle velocity in a molecular beam. By illuminating the molecular beam in the longitudinal direction, and varying the laser frequency, it is possible to excite the fluorescence of atoms belonging to different portions of the velocity distribution.

Since $\nu_{res} - \nu_{las} = \Delta\nu = \nu_{res} v \cos\theta/c$, where $\theta = \arccos(\mathbf{v} \cdot \mathbf{c})$, we easily obtain the expression for the velocity of the fluorescing group of atoms: $v = \lambda_{res} \Delta\nu / \cos\theta$. In addition to the advantages mentioned above, this detector is compact and the detection probability normalized to unit beam intensity may be up to 35%.[16]

One of the most interesting developments in the laser detection technique is the Doppler spectroscopy method proposed by Kinsey.[17] In the usual method, the laser is scanned across an absorption band and the information about internal states is extracted from the distribution of the fluorescence intensity. By illuminating the scattering center, it is possible to obtain information on the relative population of levels of all the scattered particles (total cross section). By displacing the point of illumination, it is possible to observe fluorescence due to particles within a given solid angle (differential cross section). In Kinsey's method,[17] one obtains directly the distribution over the internal states, angles of scattering, and velocities, i.e., a high-order differential cross section. This is achieved by laser scanning over the Doppler absorption line profile. Such measurements can be performed for different polar and azimuthal angles of incidence of the beam of light which can readily be varied by using fiber optics (Fig. 2c).

The usual time-of-flight method will ideally yield the

third-order differential cross section

$$\frac{d^3\sigma(v, \theta, \varphi)}{d\Omega\, dv}.$$

In the Doppler method, the measured quantity is the Doppler line profile, which is given by the integral

$$D_k(w, \theta, \varphi) = \int d^3\mathbf{v} F_k(\mathbf{v}) [\delta(w - \mathbf{n}(\theta, \varphi)\mathbf{v})],$$

where $\mathbf{n}(\theta, \varphi)$ is a unit vector in the direction of incidence of the light beam, defined by the angles θ, φ; $F_k(\mathbf{v}) = F_k(v_x, v_y, v_z)$ is the three-dimensional velocity distribution function for particles with a given internal state k, and w is the velocity component in the direction of the beam, which is related to the Doppler shift $\Delta\nu$ by the usual formula $w = \lambda_{res}\Delta\nu$.

A substantial increase in the signal-to-noise ratio can be achieved in this method because, in contrast to the small solid angle of the conventional detector, the fluorescence photons can be collected from practically the entire hemisphere. The estimated velocity resolution is ±20 m/sec (by reducing the frequency band of the laser, this resolution can be reduced down to ±0.5–5 m/sec).[17] By numerically inverting the expansion for $D_k(w, \varphi, \theta)$, it is possible to obtain $F_k(\mathbf{v})$, i.e., the most complete information about the ensemble of particles produced as a result of collisions. The full possibilities of this method can be realized only by automating the measurements.

We have already mentioned the possibility of polarizing the angular momentum of linear molecules in a beam. Fluorescence can be used to achieve selective detection of particles with a chosen direction of the angular momentum $\mathbf{J}(\mathbf{J} \perp \mu)$, since the probability W of absorption of a photon depends on the matrix element of the dipole moment μ of the molecule [$W \sim (\mu \cdot \mathbf{E})^2$, where E is the electric field in the plane-polarized wave]. This relation and the fact that, in the case of the fluorescing molecule, the probability of return to the initial state is slight, indicate that the degree of polarization of the beam can be determined by a pumping method analogous to that illustrated in Fig. 2b. The degree of polarization can be determined by dividing the laser beam into two beams, namely, a pumping and a probing beam, and by determining the fluoresence intensity with the pumping beam turned on and off. This procedure has been used[18] to obtain preliminary estimates of the degree of polarization, represented by the ratio $n_\perp/n_\parallel$ (found to be 6 for Na_2 molecules).

Laser activation, or the preparation of colliding partners in a given state, has been achieved in two ways, namely, by proceding vibrationally excited molecules and electronically excited atoms.

If the equilibrium separations in a molecule in excited and ground states are different, then for radiative times exceeding the oscillation period ($\tau_{rad} \gg \tau_{osc}$), vibrational pumping can be achieved through direct and reverse vertical Frank-Condon transitions. Radiative transitions of excited molecules result in the repopulation of the original vibrational levels.

Electronically excited targets have been described by Carter *et al.*[19] and Duren *et al.*,[20] who investigated differential scattering with Na-Ne and Na-Hg crossed beams. Continuous exposure of the sodium atoms at the point of intersection of the two beams to radiation corresponding to the transition from the $F = 2, {}^2S_{1/2}$ to $F = 3, {}^3P_{3/2}$ levels with laser output of 30–50 mW and bandwidth of 30 MHz produces an equilibrium 30% population of the excited states. As a result, the sodium beam in the illuminated region consists of a mixture of atoms in the ground and excited states, and information on the scattering of the excited particles is obtained by subtracting the known contribution due to the particles in the ground state. The macroscopic effect of excitation is seen directly in the spatial splitting of the original collimated sodium beam into two components. The absorbed photon transfers momentum to the atom and spontaneous emission, being isotropic (in contrast to stimulated emission), does not compensate this momentum. As a result of multiple absorption and emission in passing through the laser beam the atom acquires transverse momentum which is responsible for the above splitting. The measured splitting[20] is about 1 mm and is proportional to the lifetime of the atoms in the excited state. This is a potential method for measuring short lifetimes ($\tau_{rad} \leq 10^{-8}$ sec).

3. Measurement of the energy lost by particles scattered out of a monoenergetic beam through a known angle is one of the main ways of investigating inelastic atomic and molecular collisions. The change in the kinetic energy (velocity) can be reliably identified with collisional excitation or deactivation of molecular or electronic degrees of freedom. The time-of-flight method is the most effective technique for the energy range between thermal energies and several keV. In the time-of-flight method, the beam is chopped in such a way that the transmission time is much smaller than the chopping period. This type of modulation does, of course, produce a reduction in the useful intensity without a change in the background, so that there is a sharp deterioration in the signal-to-noise ratio. High-resolution measurements then require very long (often unacceptably long) signal accumulation times. This difficulty has been overcome by the correlation method of obtaining time-of-flight spectra,[21] which has long been known in neutron physics and has recently been extended to molecular beams.

In the correlation method, the source of information on the time-of-flight spectrum is the distribution obtained by periodically modulating the beam in accordance with a law determined by a binary on-off switching sequence which has a delta-shape autocorrelation function. This method has been used with both electrical[22] and mechanical[23] modulation. Figure 3 shows the appearance of the disk for the mechanical modulation of the beam.

When the beam is modulated by the above sequence, measurement of the correlation function between the detector counting rate and the modulating signal (this can be done on-line to a minicomputer; a correlation analyzer has been developed for the Vector system[24])

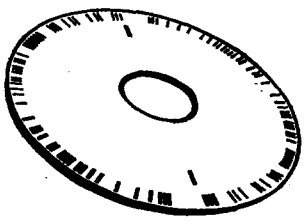

FIG. 3. Schematic illustration of disk used for correlation time-of-flight measurements with two pseudorandom sequences (103 elements) of modulating slits and obstacles. The beginning of the sequence coincides with the slit in the second row which gives start pulses due to the interruption of the light beam.

can be used to obtain a function which differs from the required time-of-flight spectrum only by a factor, so that a special measurement of the background is unnecessary. The effectiveness of the method can be understood by considering the following comparison with the single slit version: other things being equal, modulation by a sequence of N elements amplifies the time-of-flight spectrum relative to the background by a factor of $\frac{1}{2}(N+1)$ and, moreover, as the signal-to-noise ratio is reduced, the ratio of quadratic noise in the two methods tends to $\Delta_1^2/\Delta_N^2 = N/4$.

This means that, for example, when $N \geqslant 100$, the limitation on the signal-to-noise ratio mentioned above is overcome, and it would appear that measurements can be performed even under very unfavorable conditions.

Figure 4 shows the results of correlation measurements of the differential cross section with the excitation of the $\Delta j = 0 - 1$ rotational transition in the Ne-HD system.[23]

We note that the signal-to-noise ratio is 0.01. The time taken to record this spectrum was about 1.5 h, whereas reliable measurements with the same signal-to-noise ratio by the traditional method are simply impossible.

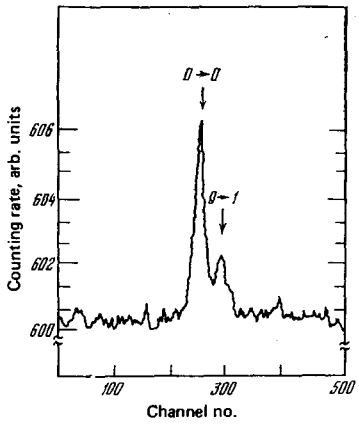

FIG. 4. Time-of-flight spectrum of a Ne beam obtained by differential time-of-flight measurements on Ne-HD crossed beams.[23] The spectrum was obtained at relative energy of 0.0303 eV and laboratory angle $\theta = 50°$. The elastic $(0 \to 0)$ peak and the inelastic peak associated with the rotational $(0 \to 1)$ transition are clearly seen.

4. It is clear from (or is implied in) the above discussion that many of the above applications are based on the utilization of a minicomputer. The automation of experiments is a topic in its own right, so that we shall only touch upon aspects of automation associated with the new techniques of detection of particles or photons through secondary-electron emission. It is well known that secondary-electron multipliers incorporating a single continuous dynode, i.e., channel multipliers, are gradually replacing the more conventional multipliers in the discrete detection of fast atomic particles with energies in excess of 100 eV and hard ($\lambda \leqslant 1000$ Å) photons.

The so-called microchannel plates,[25] consisting of a set of channels of reduced size but the same ratio of length to diameter as in the ordinary channel multiplier, are a development of these channel multipliers. A packet of this kind, prepared in the form of an integrated plate (typical diameter 30 mm, thickness 1.5 mm) may contain 10^5–10^6 independently working electron-multiplying channels per 1 cm^2, while the diameter of each individual channel is $\geqslant 10$ μ.

The amplification factor of an individual plate lies in the range 10^3–10^4 and an assembly of two plates in series can be used to obtain a large overall gain. A gain of up to 10^7 can be obtained with an assembly of this kind for a saturated (bell-shaped) distribution of pulses, which is necessary for reliable discrete particle counting. The characteristic feature of the microchannel plates is the exceptionally small length (fractions of a nanosecond) of the output pulses.

A remarkable consequence of the fact that the individual channels in the microchannel plate operate independently is that a plate of this kind can be used to decompose the image produced on its surface by a flux of particles or photons. This means that, providing one has a way of recording the amplified image produced by the microchannel plate, the system offers a position-sensitive detector with unique spatial resolution (down to 10 μ).

The simplest way of recording the image produced by the above method is to place a phosphor immediately after the microchannel plate and then inspect the image visually. This is convenient for operational control of, for example, the geometric position and size of a beam.

Figure 5 shows schematically a simple design of a position-sensitive detector with a quadrant collector,[26] ensuring electronic scanning of the image and yielding the distribution of the current density intercepted by the microchannel plate. One way of determining the coordinates of the point of incidence of the particle, which are practically the same as the coordinates of the "center of gravity" of the charge distribution of the electron shower at exit, is to divide the shower charge Q between four symmetric collectors (Fig. 5). The coordinates are then defined by the expressions

$$x = \frac{(Q_1+Q_4)-(Q_2+Q_3)}{Q_1+Q_2+Q_3+Q_4},$$
$$y = \frac{(Q_2+Q_4)-(Q_3+Q_1)}{Q_1+Q_2+Q_3+Q_4},$$

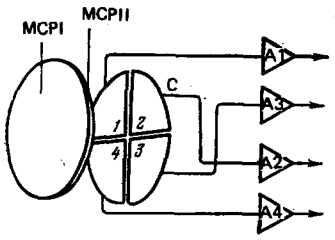

FIG. 5. Schematic illustration of the position-sensitive detector with a quadrant collector: I, II—set of two microchannel plates; C—collector, A—charge-sensitive amplifier.

where Q_i is the charge collected by the i-th collector and $x = y = 0$ corresponds to the position of the point at which the arms of the cross on the quadrant collector intersect. The best effective resolution that has been achieved in the radial coordinate is on average 10–20 μ. If we know the coordinates of the point of arrival, we can easily determine the scattering angle θ for a collimated beam ($\tan\theta = \sqrt{x^2 + y^2}/L$, and L is the distance from the center of scattering to the position-sensitive detector). Cylindrical symmetry of the scattering process of the beam enables us to achieve complete collection of the particles scattered between θ and $\theta + \Delta\theta$, i.e., speed up the measurement process very considerably. Measurement of differential scattering within the angular limits of the position-sensitive detector is thus reduced to the detection of the particle and simultaneous computation (in an on-line computer) of the coordinates of the point of impact. The result is stored in the computer which gradually accumulates the available data. A system of this kind with a fixed position-sensitive detector has been used to determine differential small-angle scattering. Figure 6 shows the results obtained for the elastic differential scattering of Li^+ ions.[26] This curve consists of about 200 points and typically requires about 10 min of measurement. This technique reveals interesting possibilities for the investigation of small-angle differential scattering, for example, in charge transfers, elastic and inelastic scattering of fast beams, and in the method of parallel beams. Detecting systems with space-time high-resolution analysis based on the position-sensitive detector appear to be very promising indeed.

Other methods of electronic reading of the information contained in an image have also been reviewed in the literature (see the bibliography in Ref. 26) but have not as yet been adopted in experiments with beams.

A possible unforeseen obstacle to the application of the microchannel plate as a position-sensitive detector is the complexity of the electronics that has to be used, on the one hand, and the possible relatively high background, on the other.

One way of resolving these difficulties can be based on the suggestion put forward by Arikava[27] and usually referred to as Hadamar spectroscopy (Ref. 28). This method of reading the image is based on mechanical scanning but, instead of the usual narrow slit, one uses a mobile multislit mask which completely covers the image. Successive displacement of the mask in front of the detector through N positions (equal to the

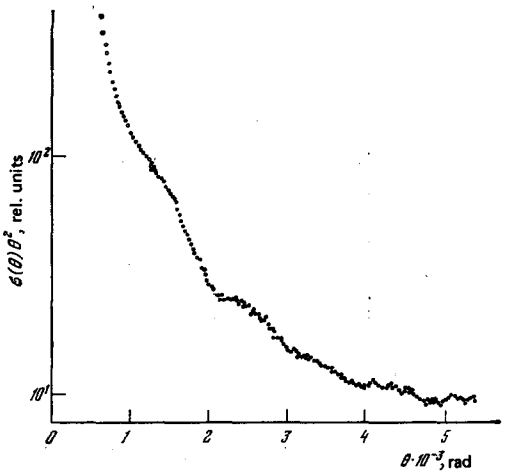

FIG. 6. Typical differential curve obtained with a fixed position-sensitive detector.[26] Li^+ beam of 600 eV, scattered by air.

number of standard slots and obstacles) gives N measured counting rates which, after decoding, can be used to reconstitute the true spatial distribution of the flux intercepted by the input surface of the detector. Thus, instead of successively measuring N spectral elements of the (one-dimensional) image, Hadamar spectroscopy measures N linear combinations of these spectral elements. The optimum method of combining the elements, i.e., of distributing the slots and obstacles, is found from the condition of minimum noise.

The decoding of measurements with the aid of the Hadamar transform results in an improvement by a factor of $(N+1)/2\sqrt{N}$ in the signal-to-noise ratio, similarly to the improvement discussed in connection with correlation time-of-flight measurements. The efficacy of this approach in the case of H_3^+ beams has recently been demonstrated in Ref. 27.

The new methodologic developments discussed above in relation to molecular-beam work undoubtedly deserve careful attention. They provide a way of substantially expanding both the range of molecular-beam research and the volume of the data obtained thereby.

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