05320-6004-R000

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November, 1966

Technical Report

Prepared Under

Contract NASw-1336

For

National Aeronautics and Space Administration

Headquarters

Washington, D.C. 20546

TRW SYSTEMS

One Space Park, Redondo Beach, California

*This work supported by the NASA under Contract NASw-1336.

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ABSTRACT

Ionization probabilities, β, have been measured for iron atoms incident upon target gases of He, Ne, N_2, CO_2, and air. The energetic Fe atoms are obtained by injecting solid iron particles of known velocity and mass into a low pressure gas target where collisional heating raises the temperature of the particle to the vaporization point. Atoms evaporated from the particle traverse the gas at a velocity essentially equal to the velocity of the solid particle and these atoms constitute the atomic "beam". For velocities in excess of $\sim$ 20 km/sec the incident particle is completely vaporized and the number of atoms injected into the target gas, N_A, is specified by the particle mass. The number of ions produced in the target volume, N_I, is determined by means of a parallel plate ionization chamber. By definition, $\beta = N_I/N_A$. An estimated value of the ionization cross-section σ_I can be obtained from these results and for the particular case of Fe atoms incident on a N_2 target, the data are in qualitative agreement with published data obtained using more conventional techniques.

*This work supported by the NASA under Contract NASw-1336.

1. INTRODUCTION

A recent paper by two of the authors[1] described a method of measuring the ionization probability, β, for certain types of metallic atoms incident on gaseous targets and data were presented for iron atoms interacting with target gases of air and argon. Additional measurements utilizing somewhat improved techniques have been completed and the results are reported herein. In the present work, β was determined for Fe atoms incident on target gases of He, Ne, N_2, CO_2, and air over the velocity range from about 15 km/sec to about 45 km/sec. This corresponds to an energy range from 65 eV to 590 eV for the incident Fe atoms.

The ionization probability as used here is defined as the ratio of the number of ion-electron pairs N_I created in a thick gas target to the number of atoms N_A injected into the gas target at a given velocity or energy. The term "thick" implies that the target gas density and volume are large enough to ensure thermalization of the most energetic atoms while within the confines of the target volume. It can be seen that β is not a true probability since it can exceed unity for high incident energies. In the limit of a single collision, β is equal to the ratio of the ionization cross-section, σ_I, to the collision cross-section, σ_C, since β is the probability that a given collision will produce an ion pair. For multiple collision conditions, β becomes a total ionization probability and is equal to the sum of the ionization probabilities for each collision involved in thermalizing the incident atom.

If the incident atom energy lies in a region where the ionization cross-section falls off rapidly with decreasing energy, the first collision is the only one which contributes significantly to the sum. In this case the ionization probability is, to a good approximation, simply the ratio of cross sections mentioned above. When the ionization cross section is not changing rapidly with energy, collisions other than the first contribute and β is a more involved function of the cross sections. β represents an upper limit to the ratio of ionization cross section to total cross section. It is possible to estimate σ_I from the experimental data, and for the particular case of Fe atoms incident on N_2 molecules, the

data is shown to be at least in qualitative agreement with the results obtained by Bukhteev and Bydin.[2]

2. EXPERIMENTAL PROCEDURES

A detailed description of the basic experimental method is contained in Ref. 1 and only a brief review will be given here. The energetic Fe atoms are obtained by introducing high velocity, sub-micron diamter, solid iron particles into a low pressure ($\sim$ .05 to 0.2 Torr) target gas. The solid particle is heated and vaporized by collisions with the gas molecules in the target gas. The particles are obtained from an electrostatic particle accelerator[3] which consists principally of a 2-million-volt Van de Graaff generator and a charged particle injector.[3,4]

A sketch of the experimental assembly is shown in Figure 1. The velocity v_o and charge q of each particle is measured prior to entering the gas target by means of capacitive detectors which have been described elsewhere.[4,5] The signal from one of these detectors, which is in the form of a rectangular voltage pulse, is displayed on one trace of a dual beam oscilloscope (Type 555 Tektronix). Although the output signal from an individual detector is sufficient to specify the particle velocity, the accuracy is improved by providing a longer flight path. In practice, the oscilloscope sweep is triggered by the output signal from a separate detector upstream from the charge detector. The time delay between the start of the oscilloscope sweep and the appearance of the charge detector signal is simply the transit time between the detectors which specifies the velocity. The function of the particle parameter selection system[6] shown in Figure 1 is to select particles within a velocity range compatible with a given set of conditions. All others are automatically deflected from the experiment.

Given particle charge q and velocity v_o from analysis of the data and the total accelerating voltage V, the particle mass m_o is calculated from the conservation of energy equation i.e., $1/2\ m_o v_o^2 = qV$.

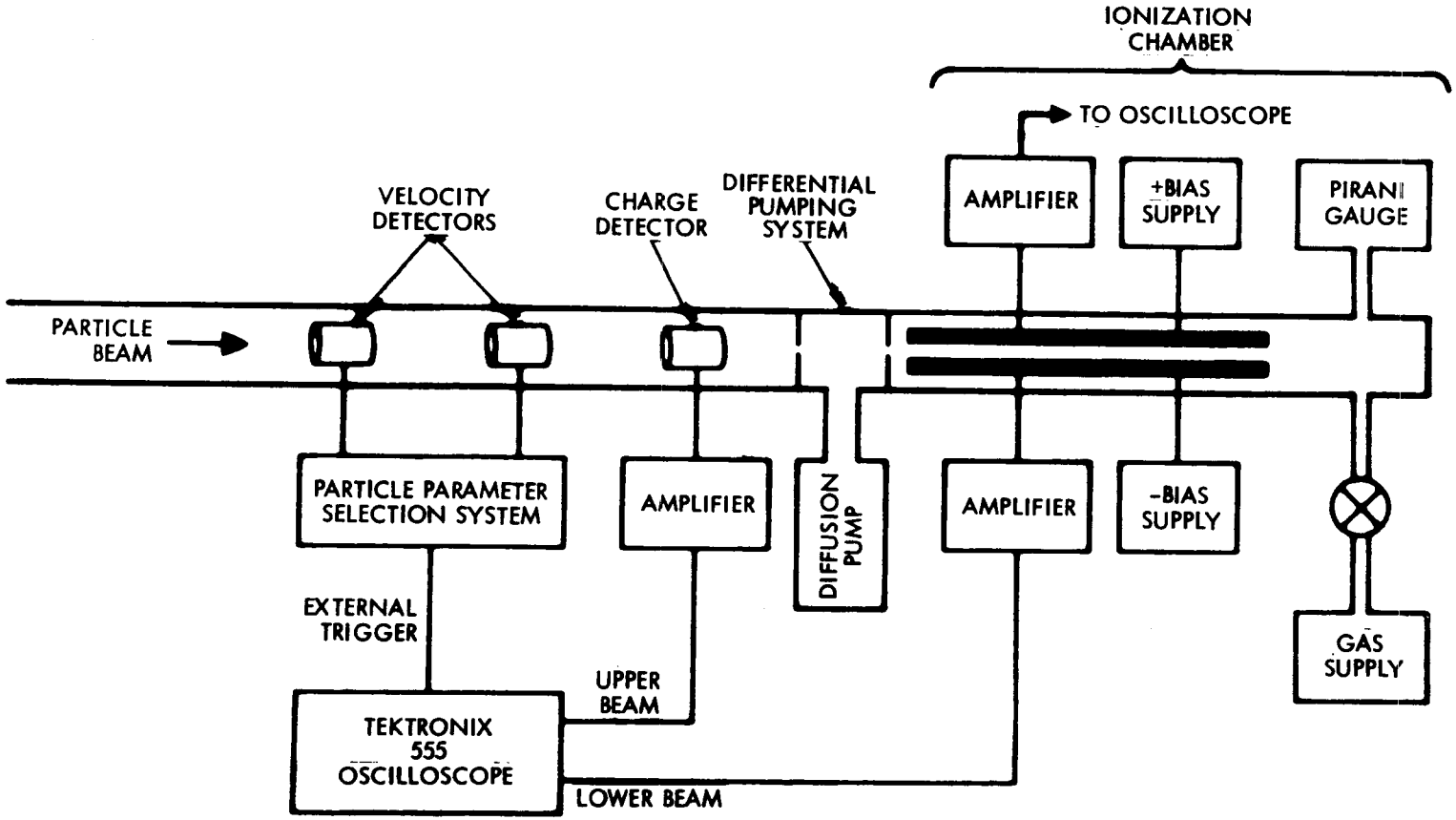

Figure 1. Schematic diagram of experimental apparatus.

The solid particles are injected into the gas target region through the channels of a one-stage differential pumping system. The pressure within the target chamber is adjusted by means of a precision variable leak control and is continuously monitored with a Pirani gauge. The absolute pressure of the target gas is not particularly critical; the principal requirement being that the particle be completely vaporized while within the confines of the target chamber.

Upon entering the target gas, the temperature of the solid particle is raised to the vaporization point due to collisions with target gas molecules. It can be shown that the remaining mass of an evaporating particle which is decelerated from an initial velocity v_o to a lower velocity v is given by

$$m = m_o \, e^{-\frac{\sigma}{2}(v_o^2 - v^2)} \tag{1}$$

where $\sigma = \lambda/2\Gamma\zeta$. The quantities λ, Γ, ζ are, respectively, the heat transfer coefficient, the drag coefficient, and the latent heat of vaporization. As shown in Ref. 1, to obtain $m = .01\, m_o$ requires only a 4% decrease in velocity for an initial velocity of 40 km/sec. In other words, for high initial velocities, where the kinetic energy per atom is large compared to the binding energy, the particle is essentially totally vaporized while suffering negligible (or at least small) deceleration. Thus, the number of atoms injected into the target gas is simply the total number of atoms contained in the solid particle.

The velocity of atoms evaporated from the surface of the particle relative to the particle is presumed to be governed by the surface temperature of the particle ($\sim$ 3,000°K). The mean thermal (random) velocity is quite small compared to the translational velocity of the particle and, therefore, the atoms have a directed velocity very nearly equal to the initial velocity of the solid particle. The pressure within the target chamber is such that the collisional mean free path of atoms evaporated from the particle is very large (by a factor of about 5,000) compared to

the particle diameter. Even though the velocity of the evaporated atoms with respect to the particle is relatively small, the probability of a vaporized atom or one of its reaction products having a subsequent collision with the primary particle is small.

Having specified the total number of atoms injected into the gas and their mean velocity, the remaining part of the problem is to measure the number of ion-electron pairs created in the gas. This is accomplished by means of an ionization chamber consisting of a pair of gold-plated parallel plates 56 cm long by 8.8 cm wide separated by a distance of 5.3 cm. The particles are injected along a plane midway between the two plates. The two plates are electrically biased in opposite directions; one serving as the positive ion collector and the other collects electrons (or negative ions). Each of the plates is a-c coupled to high input impedance solid state amplifiers. The amplifiers have an overall voltage gain of about 50 and a pass band from 100 cps to 1 mc/sec. The RC decay time of the input stage was fixed at about 1/2 msec by adding a fixed capacitance in parallel with the capacitance to ground of the collector plate. Since the RC decay time is very long compared to the time interval over which ions (or electrons) were collected, the ion (electron) current is effectively integrated at the input stage. The total charge collected Q_c is given by $Q_c = V_c\ C_c/G$ where C_c is the total input capacitance of the amplifier, V_c is the amplitude of the integrated signal in volts, and G is the voltage gain of the amplifier. Assuming singly ionized atoms, the number of ions created N_I is equal to Q_c/e where e is the unit electronic charge. The method of measuring C_c and G has been described in Ref. 1. The output signals from the two collectors are displayed on oscilloscopes and photographed for later analysis. Correlation of the output signals with the parameters of the particle which created the ions is accomplished in a straightforward manner.

The principal improvement over the work reported in Ref. 1 has been in the design of the ionization chamber. Two opposing problems, which can effect the validity of the results, combine to place somewhat stringent requirements upon the absolute gas pressure, chamber geometry, and ion collection voltage. The problems are electron multiplication in the gas

and the escape of energetic ions and atoms.

To avoid electron multiplication, which would increase the number of ion pairs created in the gas target, a low electric field and a short collisional mean free path are required. However, if the electric field is decreased by lowering the potential bias on the collecting plates, it becomes possible for scattered ions to reach the positive collecting plate instead of the negative plate. This problem is most severe at high energies and, even after several collisions with gas molecules, the ion energy may be sufficiently great to overcome the potential barrier at the plate.

One way of avoiding this problem is to increase the absolute gas pressure so that the ions suffer more energy absorbing collisions before reaching the wall. This has the added virtue of lowering the probability of electron multiplication. However, increasing the target gas pressure decreases the range of the primary particle. Since the differential pumping system has channels of finite length (1 cm), the particle begins to heat up within the channel and, for excessive pressures, particle vaporization can begin before the particle reaches the main target chamber. Ion pairs produced in this region would not be collected.

Although there is no unique solution which satisfies all of the requirements over the entire energy range, the target chamber was designed with these considerations in mind. Sufficient latitude was available to minimize the effects of ion escape and electron multiplication for any given set of conditions. The existence of either of these effects could be determined by detailed examination of the signals from the ionization chamber.

In practice, the collector voltages were kept as small as possible consistent with negligible ion loss while simultaneously maintaining the highest gas pressure possible consistent with the problem of premature particle vaporization. Possible errors due to these effects are estimated to be less than a few per cent at high velocities and are negligibly small at the low end of the velocity range. The actual value of the collector voltages ranged from plus and minus 45 volts to plus and minus 11 1/2 volts

depending upon the target gas and the velocity range under study. In retrospect, it appears that the ion escape problem may have been underestimated to some extent in the earlier paper because some discrepancy has been noted for air data obtained with the different target chambers, particularly at the high velocity extreme. However, the probable errors calculated from the least squares fit to both sets of data overlap even at high velocities. At low velocities where ion escape is negligible, the data are in excellent agreement.

3. EXPERIMENTAL RESULTS

The experimental results for Fe atoms incident on target gases of He, Ne, N_2, air, and CO_2 are shown in Figures 2 through 6, respectively. Each point represents the data obtained from an individual particle and gives β as a function of the initial particle velocity. No correction was made for the deceleration suffered by each particle. The number of atoms per particle N_A ranges from about 10^8 at 20 km/sec to about 10^7 at 45 km/sec. The absolute number of ions collected varied from about 3×10^5 to 3×10^7 depending upon the velocity, target gas, and β. As noted previously, the solid particles are positively charged when they enter the gas target. This charge is eventually collected by the ion collector. However, no correction was made to the data since the number of unit electronic charges carried by the particle (typically 10^3 to 10^4) is negligible compared to the number of ion-electron pairs formed in the gas. N_I was determined from the total positive charge collected at the negative collector. As an internal check for consistency the total negative charge collected at the positive collector was also monitored and, as expected, the results were identical. It should be pointed out that no attempt was made to discriminate between electrons and negative ions collected at the positive plate. For most of the target gases, the probability of negative ion formation is probably negligible with the possible exception of air and CO_2. Bukhteev, et al.,[7] have measured negative ion formation cross sections for the cases $K \rightarrow O_2$, $K \rightarrow Cl_2$, and

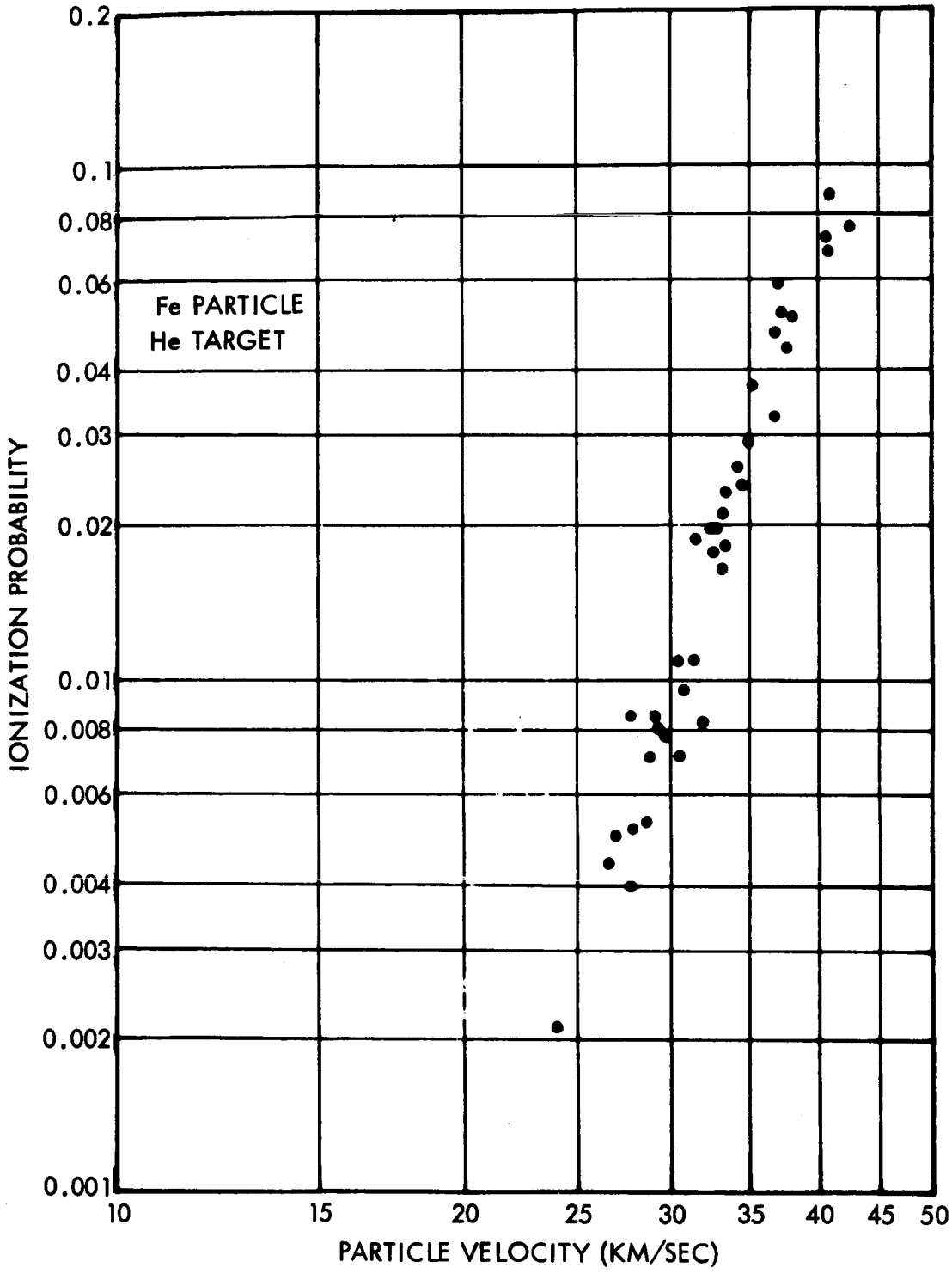

Figure 2. Ionization probability, β, as a function of particle velocity for iron particles and a helium target.

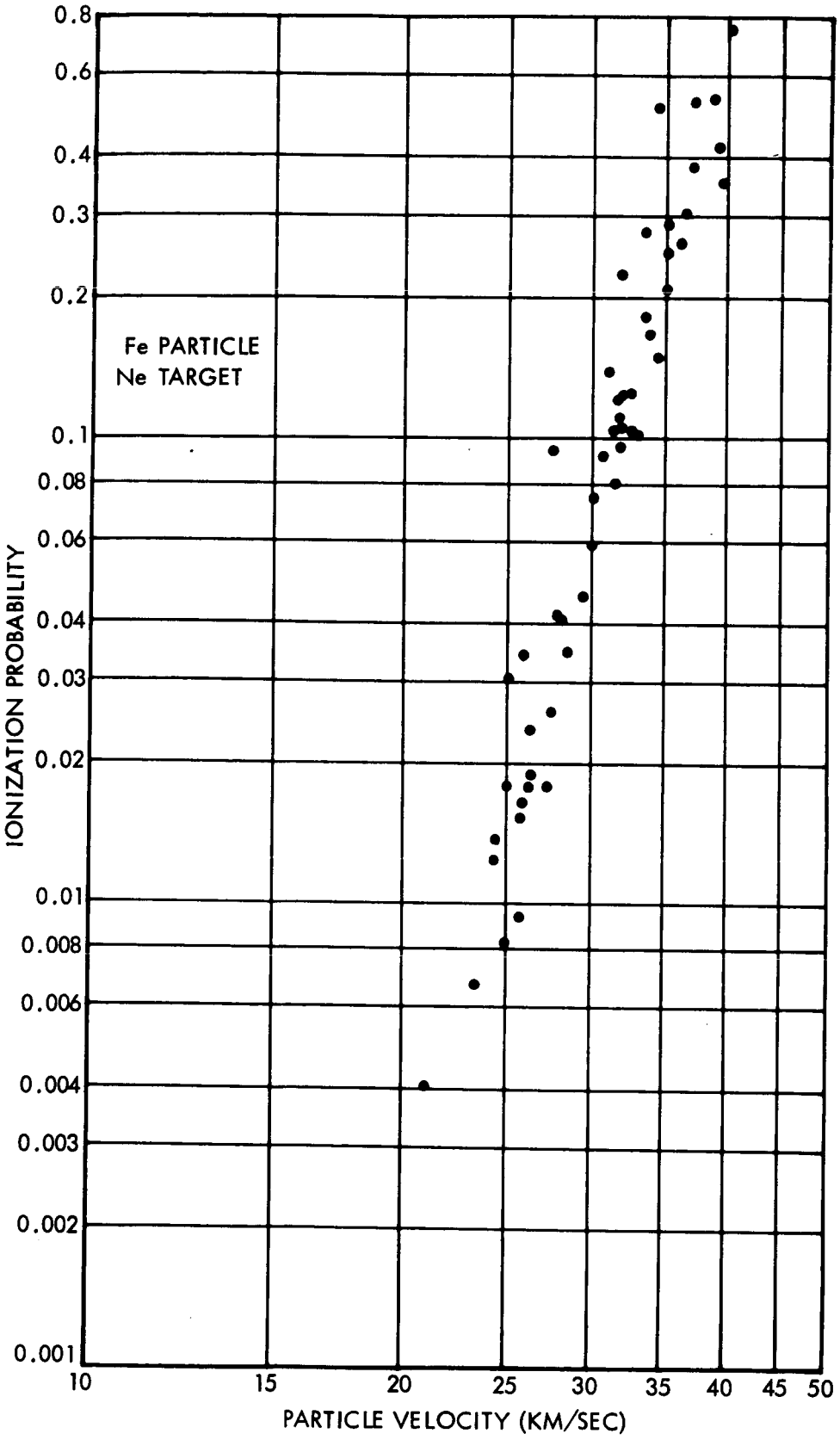

Figure 3. Ionization probability, β, as a function of particle velocity for iron particles and a neon target.

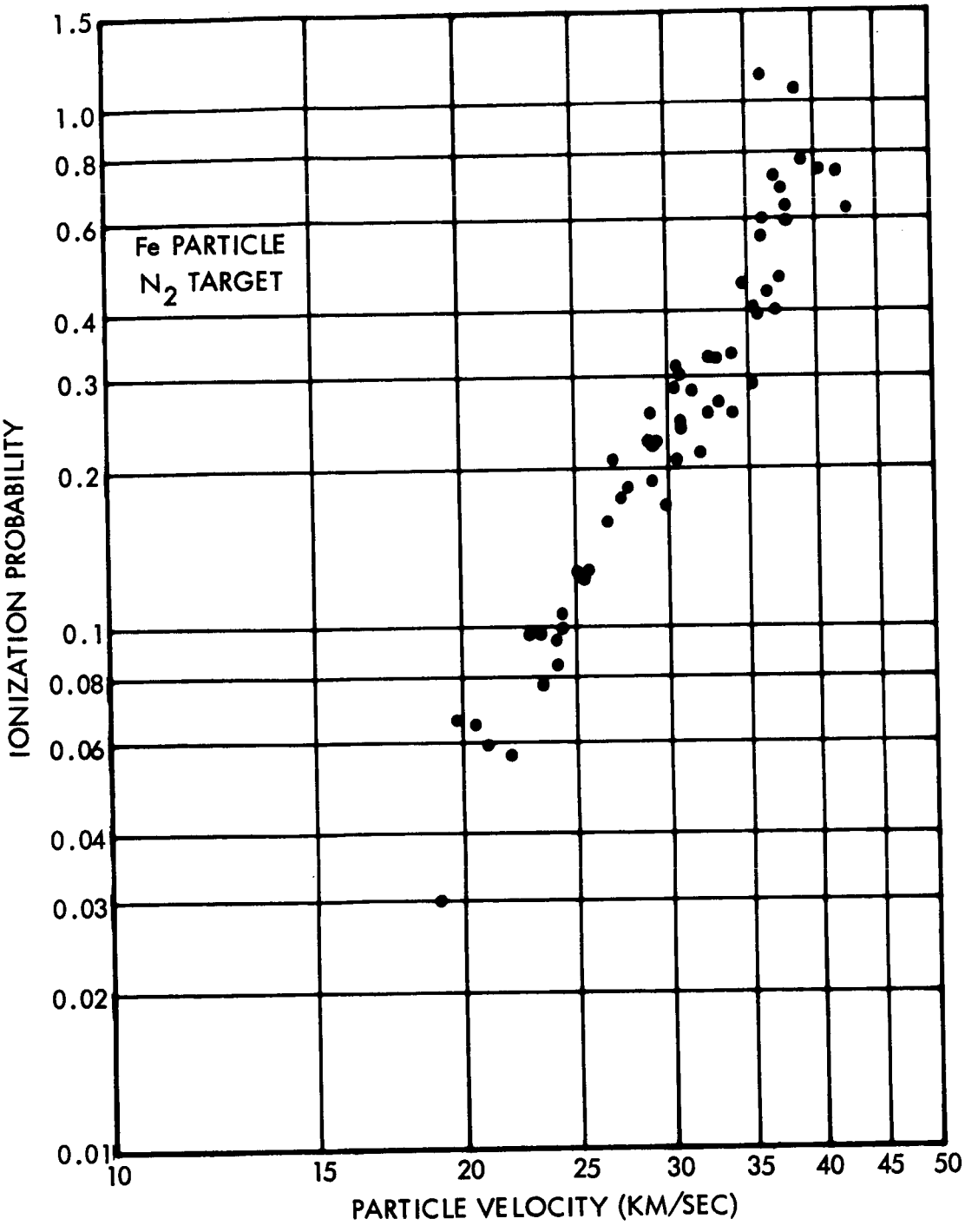

Figure 4. Ionization probability, β, as a function of particle velocity for iron particles and a nitrogen target.

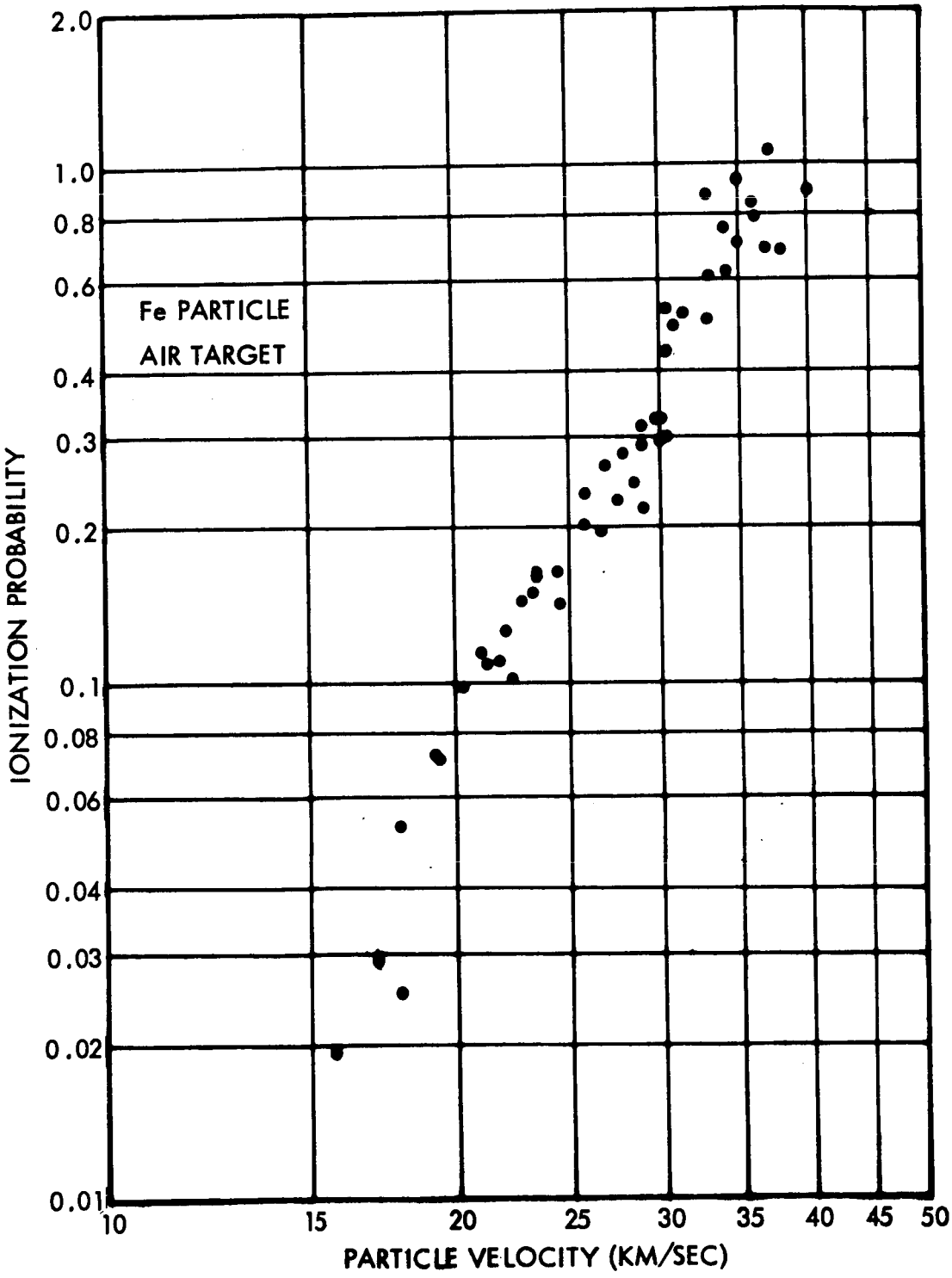

Figure 5. Ionization probability, β, as a function of particle velocity for iron particles and an air target.

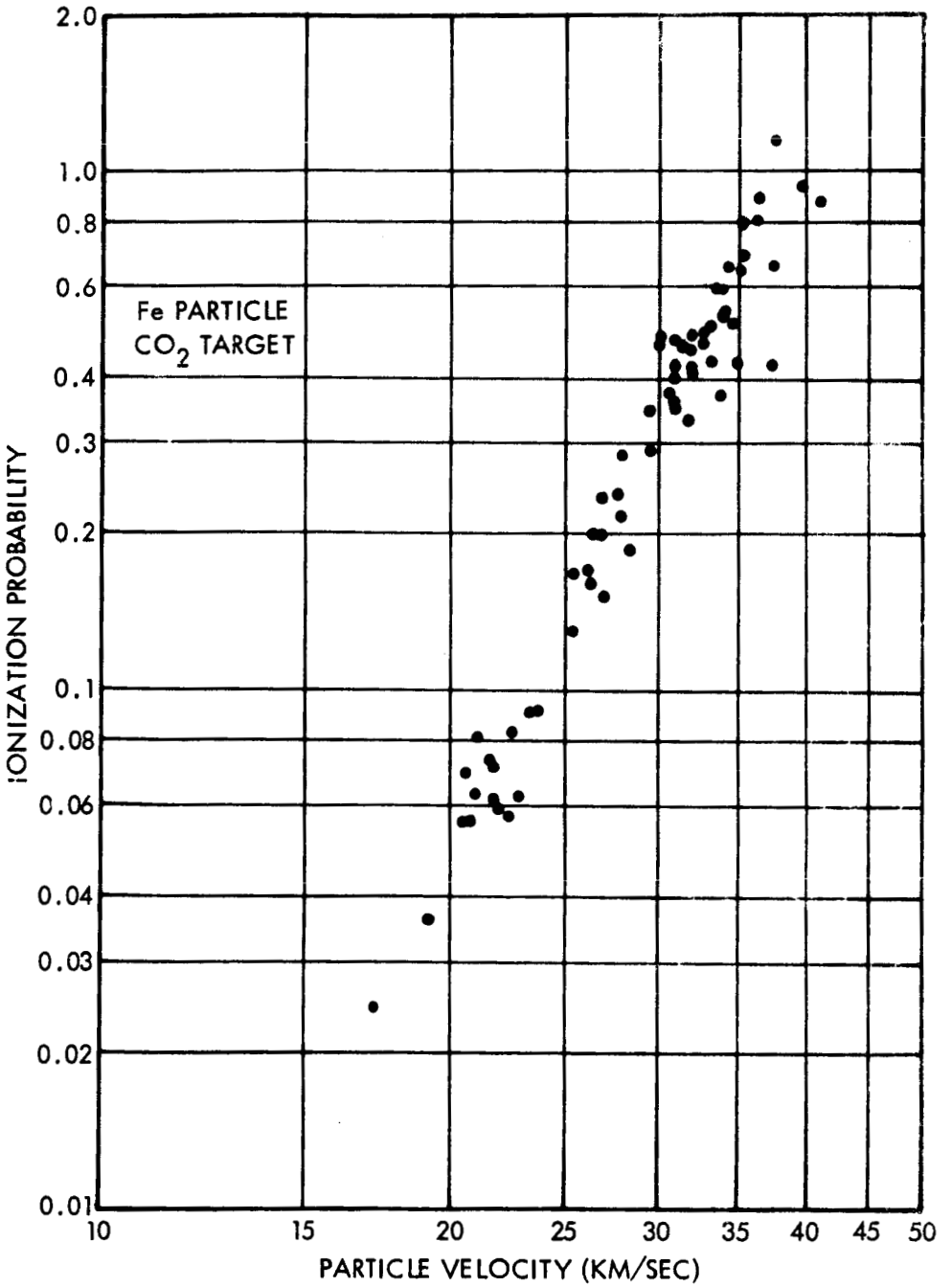

Figure 6. Ionization probability, β, as a function of particle velocity for iron particles and a carbon dioxide target.

Rb → O_2 and, at low energies, this is the preferred process.

The principal source of random error in these experiments is in the measurement of initial particle charge, q. Typically, signal-to-noise ratios for signals from the charge detector range from 3 or 4 to one at high velocities to better than ten to one at the low velocity end. The probable error in determining q ranges from an estimated $\pm$ 15% at high velocities to $\pm$ 5% at low velocities. Random errors in the measurement of v_o are probably less than $\pm$ 3% and the accelerating voltage is known to about $\pm$ 1%. All of these quantities combine to effect the accuracy of the measurement of N_A. Random errors associated with the measurement of N_I are much less, totaling an estimated $\pm$ 3% due principally to reading the data from the photographic records.

Possible systematic errors occur in the measurement of the capacitance and gain of the amplifiers used on the particle charge detector and ion collector. The absolute calibration of the accelerating voltage also must be considered. It is estimated that the total systematic error due to these effects is less than $\pm$ 18% over the entire range of velocities. A great deal of care was exercised in minimizing ion escape and electron multiplication with the result that errors due to these effects are considered to be negligible at low velocities and perhaps as much as $\pm$ 10% at the extreme high velocity end.

In order to draw a comparison between the effects of the target gas on β, free hand curves representing the data of Figures 2 through 6 are shown in Figure 7. An energy representation is also commonly used. Since the kinetic energy of the center of mass is a constant of the motion, the energy available E_A for inelastic processes is the difference between the kinetic energy of the moving particle in the laboratory system and the kinetic energy of the center of mass. Assuming that the Fe atom is preferentially ionized because of its low ionization energy (E_I = 7.85 eV), β is plotted as a function of $E_A - E_I$ in Figure 8 for the same curves given in Figure 7. We have not experimentally verified the assumption that the Fe atoms are preferentially ionized.

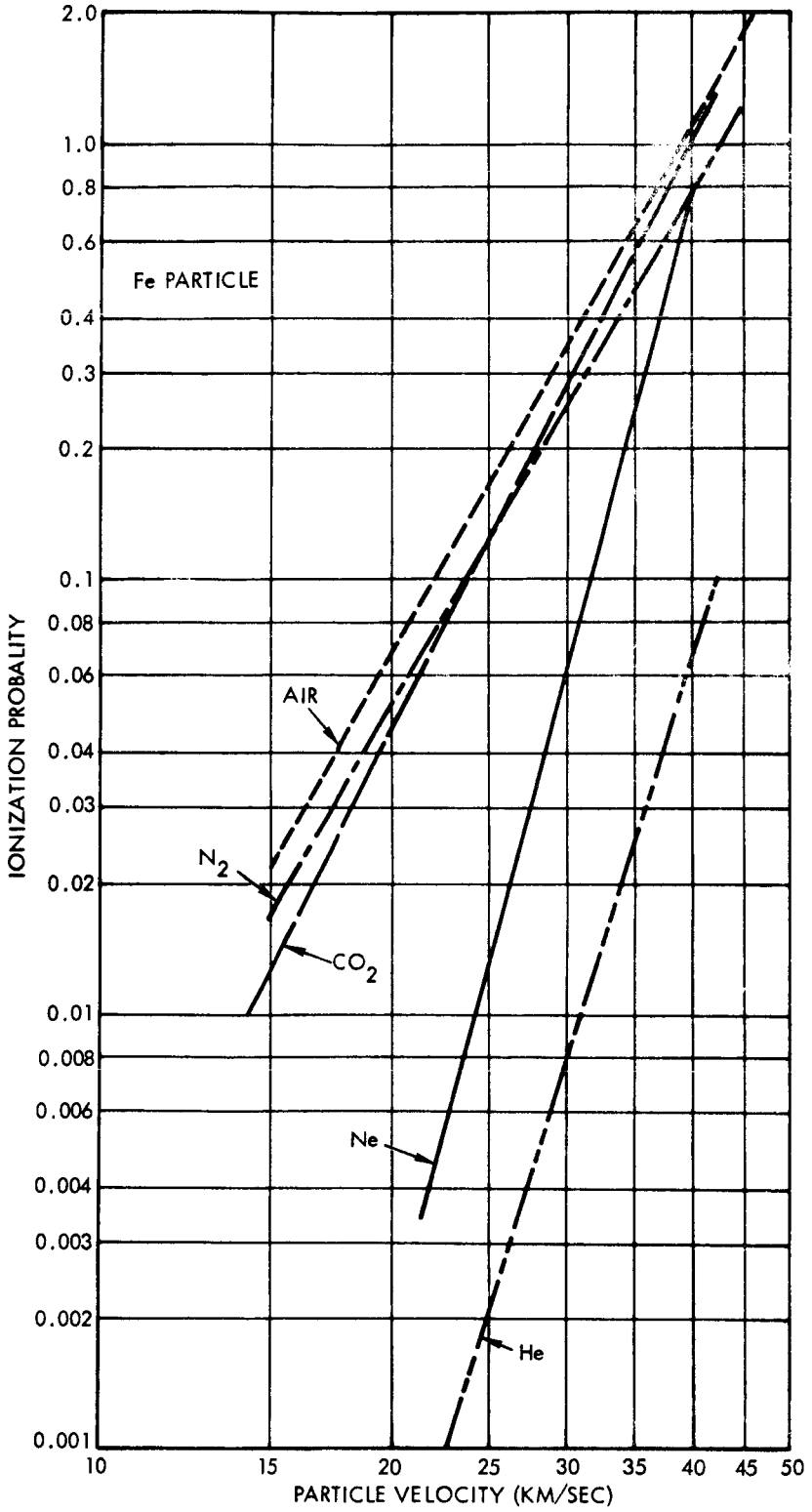

Figure 7. Ionization probability, β, as a function of incident particle velocity for iron particles and several different gases. The lines represent the best free hand fit to the data given in Figures 2 through 6.

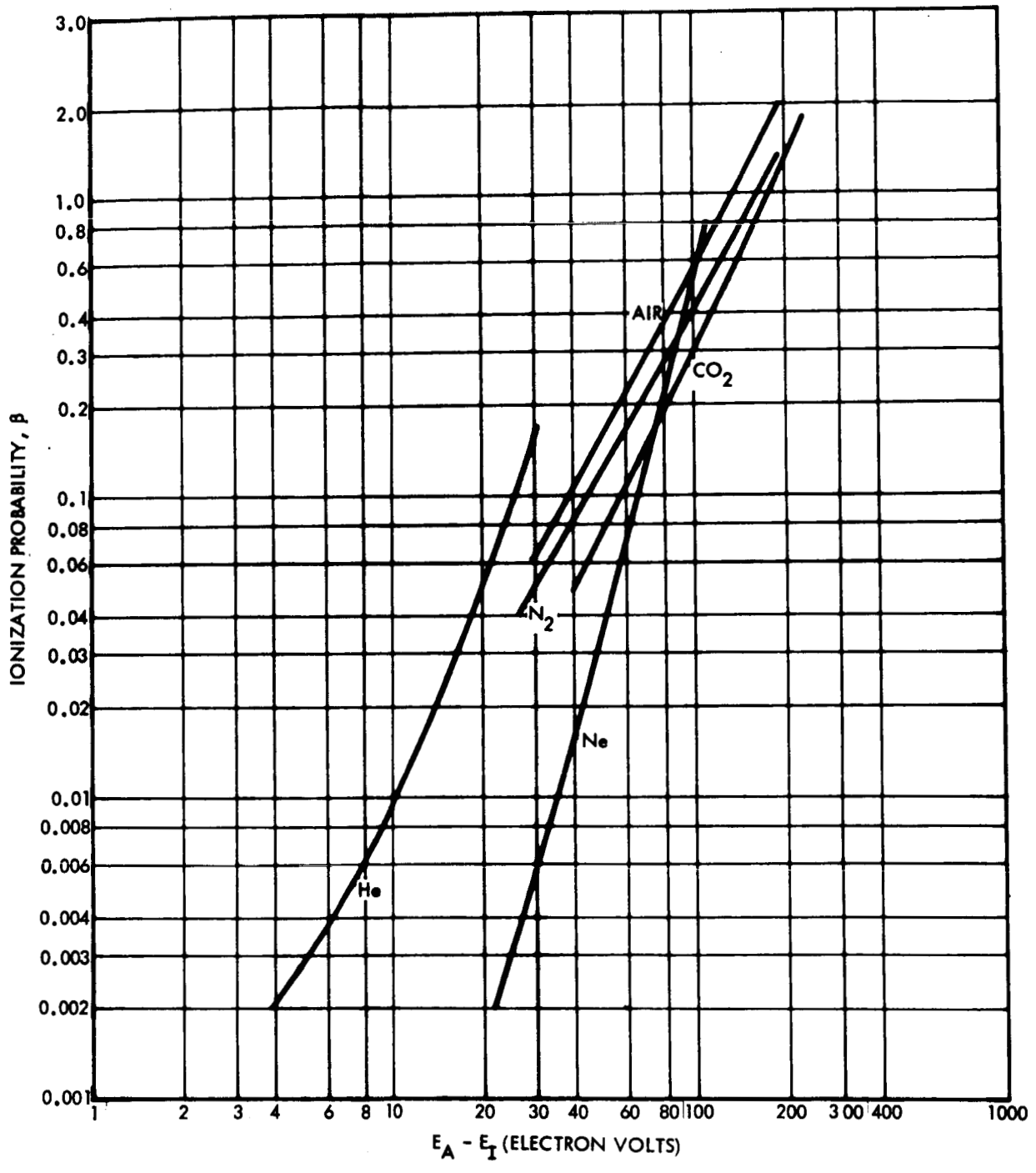

Figure 8. Ionization probability, β, as a function of energy in excess of the ionization energy for iron particles and several target gases. The lines represent the best free hand fit to the data given in Figures 2 through 6.

4. DISCUSSION OF RESULTS

Ionization processes are usually described in terms of the ionization cross-section σ_I rather than β. In molecular beam work, cross sections are determined by using thin targets where the probability of multiple ionizing collisions is negligible. For the present work this is not the case. We require that the target gas collisions provide the energy to vaporize the particle and the target gas pressure and volume are fixed by this requirement. In all cases, the vaporized atoms and reaction products suffer multiple collisions in the gas. In principle, σ_I can be calculated from β, but this requires a detailed knowledge of scattering processes, etc., which is not presently available. However, an estimate of σ_I can be made and, as shown below, the results agree with those obtained by other experimenters.

Consider the case of Fe atoms incident on N_2 molecules (molecular weight $\sim$ 28). Assuming isotropic scattering in the center of mass system, the average kinetic energy of Fe atoms following a collision is about one-half of the initial kinetic energy of the atom. Let E_A represent the energy available for inelastic processes at the first collision of an ion atom with a N_2 molecule. After the first collision the energy available is 1/2 E_A. Now if the initial energy is such that 1/2 $E_A \lesssim E_I$ we can safely assume that σ_I for the second collision is negligibly small. In this idealized case, then, the measured β is essentially equivalent to σ_I at the energy corresponding to the initial energy of the incident Fe atom. By this logic, only a small number of ionizing collisions are energetically possible. An analysis of this kind indicates that an estimated 50 to 80% of the ion pairs produced at the highest energies available are due to the first collision of the incident atom with a gas molecule. At lower energies the fraction of first collision ionizing events is probably higher.

Another factor that should be considered is the possibility of ionization from excited states due to multiple collisions. In order for this to be a major effect, very large excitation cross sections would be required and it is assumed that this type of stepwise ionization is small compared to direct ionization from a single collision. Thus, to a first

approximation at least, $\sigma_I = \eta\beta\sigma_c$ where σ_c denotes the collision cross section and η varies between one-half and one depending upon the initial energy of the Fe atom.

The only experimental results available to which a direct comparison may be made are those of Bukhteev and Bydin.[2] Using an atomic beam technique, they determined ionization cross sections for Fe atoms incident on a N_2 target gas. They give values of $\sigma_I = 0.25 \times 10^{-16}$ for an Fe atom velocity of 80 km/sec and $\sigma_I = 0.4 \times 10^{-16}$ at 100 km/sec. They point out that these are only order of magnitude results because ions scattered through angles larger than 1° 48' were not detected. We assume that they have not made a large angle scattering correction to their data. Assuming that the scattering function is isotropic in the center of mass system leads one to believe that their measured cross section are, to within an order of magnitude, equal to the collision cross section, σ_c. From our work $\sigma_I \simeq 0.6\ \sigma_c$ for Fe → N_2 reactions at a velocity of 40 km/sec. Here we have assumed $\eta = 0.7$ and, from Figure 7, $\beta = 0.8$. Thus, the results are in good agreement, at least to the extent that the approximations made are valid.

On a more qualitative basis, some additional generalizations may be drawn from the experimental results. For example, the data presented in Figures 7 and 8 indicate that the rate of change of β with velocity (or energy) appears to be systematically greater when the target gas is monatomic (He and Ne) than is the case for diatomic (N_2 and air) or triatomic (CO_2) target gases. Also, the ionization cross-sections estimated from our results are significantly greater than those obtained when both the target and projectile molecules are common gases. For example, a number of recent papers,[8-12] have given the results of ionization cross section measurements with most combinations of H_2, He, Ne, O_2, N_2, and A molecules. Our results appear to be similar to those of Bydin and Bukhteev[13] who obtained σ_I for alkali metal atoms interacting with a variety of target gases. The apparent conclusion is that metal-like atoms have large ionization cross sections.

5. SUMMARY

The method of producing an "atomic beam" by vaporization of solid metallic particles has been used to determine ionization probabilities for Fe atoms interacting with various gas targets. The results obtained appear to be compatible with those obtained by more conventional techniques. Except for the problem of determining σ_I from the measured β, the experimental procedures are quite straightforward and the magnitudes of the experimental probable errors are smaller than those usually stated for atomic beam work when similar types of projectile atoms (i.e., metallic) are used. The range of energies available is limited at the lower end by the requirement of total particle vaporization and at the upper end by the characteristics of the particle accelerator. The choice of particle materials is limited to metals (or at least relatively good conductors), but, the use of this technique promises to increase the number of ionization reactions which may be studied experimentally.

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