

THE PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, VOL. 128, No. 4

NOVEMBER 15, 1962

Theory of Negative Resistance Characteristics in Irradiated Gas Thermionic Diodes

R. FORMAN

Parma Research Laboratory, Union Carbide Corporation, Parma, Ohio

(Received July 13, 1962)

A theory explaining negative resistance effects in the anode current-voltage characteristics of inert gas-filled thermionic diodes exposed to ionizing radiation is proposed. The theory shows that these effects are observed in argon-, krypton-, and xenon-filled diodes under appropriate conditions of anode spacing, gas pressure, and radiation dosage because these gases have a Ramsauer minimum in their electron scattering cross-section data. The theory also shows that gases not having this anomaly in their scattering cross section, such as neon or helium, do not show negative resistance effects.

INTRODUCTION

EXPERIMENTAL data on inert gas-filled thermionic diodes have shown that diodes containing the gases argon, krypton, and xenon have negative resistance characteristics, whereas helium- and neon-filled tubes do not have this property.¹ An obvious difference between these two classes of gases is that the former has the Ramsauer effect² and the latter does not. The Ramsauer effect leads to an electron scattering behavior in the gas such that at very low electron energies the mean free path increases with increasing energy, and at higher energies the mean free path decreases with increasing electron energy. This change in scattering cross section in the region of 1 eV has been proposed previously to explain the negative resistance phenomena observed in fission product krypton diodes.³

In order to place these speculations on firmer theoretical grounds, an attempt was made to apply the theory of hot electrons⁴ to the problem. Adawi,⁵ using a modification of this theory, has shown that negative resistance effects similar to those observed in irradiated inert gas diodes are unlikely in a gaseous discharge. To obtain this result, he assumes a model in which the mean

free path monotonically increases with electron energy. Attempts were made to show that Adawi's theory would lead to negative resistance effects if the mean free path through the gas was assumed to have a Ramsauer maximum, but these trials were not successful. As a result, it was concluded that the hot electron approach⁴ to the problem was inapplicable for two reasons: (a) It assumes the electrons attain thermal equilibrium with the gas and does not consider the possibility that the hot electrons could pick up practically all the energy of the applied voltage, and (b) it did not take into consideration the finite distance between emitter and collector in the plasma. The first condition could arise because of the infinitesimal loss of energy in an elastic collision between an electron and inert gas atom and the fact that low-energy electrons have a long mean free path in xenon, krypton, and argon. These two characteristics, in conjunction with the finite distance between emitter and collector (condition b), can lead to a situation where the electron picks up more energy from the field per unit time than it loses by collision and reaches the anode collector with almost the full energy of the applied voltage. To illustrate a consequence of this situation mechanistically, consider Fig. 1. Figure 1(a) shows the model considered. The cathode and anode are separated by a plasma containing a predominance of neutral particles G which are the elastic scattering centers. A variable voltage V accelerates electrons in the space. Figure 1(b) shows a typical Ramsauer effect curve where the mean free path (λ) is plotted as a function of electron energy (ϵ) for low values of electron energy. If

¹ R. Forman, J. A. Ghormley, and J. R. Reiss, following paper [Phys. Rev. **128**, 1493 (1962)].

² C. Ramsauer, Ann. Physik **64**, 513 (1921).

³ R. Forman and J. A. Ghormley, J. Appl. Phys. **33**, 3057 (1962).

⁴ S. Chapman and T. G. Cowling, *The Mathematical Theory of Nonuniform Gases* (Cambridge University Press, New York, 1960), 2nd ed., p. 346-352.

⁵ I. Adawi, J. Appl. Phys. **32**, 1101 (1961).

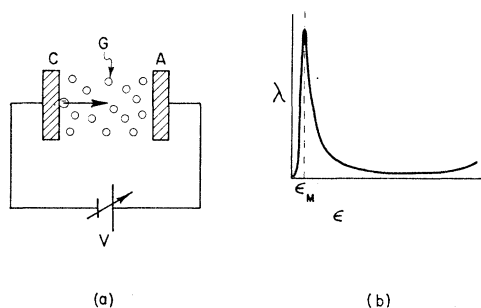


FIG. 1. (a) Simple model for gaseous diode. C is the cathode, A is the anode, and G is the neutral gas particles. (b) Mean free path vs electronic energy data for a typical gas (e.g., xenon, krypton, argon) having a Ramsauer minimum in its electron scattering cross-section data.

the pressure of the gas G and the distance between C and A are such that an electron can continually gain energy from the electric field as described above, a number of different conditions can arise depending on the value of V . If V is less than ϵ_m , an electron emitted from C will appear to see a less dense gas (longer mean free path) as it accelerates until it is collected at A . When V is greater than ϵ_m , then the electrons from C initially appear to see a less dense gas as they progress through the space until they attain an energy ϵ_m . At this point in space the previous situation changes and the electrons seem to see a denser gas as they progress toward the anode. It is obvious that this change in scattering mechanism could lead to a situation where the transit time for an electron would be longer at a higher applied voltage than at a lower value of V . Under these circumstances it is possible to obtain negative resistance.

On the basis of the conceptual ideas described above, a simple theory has been devised which is in substantial agreement with experimental data obtained in irradiated inert gas diodes. This theory shows that the negative resistance measured in xenon, krypton, and argon irradiated diodes is a consequence of the Ramsauer minimum in the electron collision cross section of these gases.

THEORY

The general features of the proposed model (planar case) are illustrated by Fig. 2. The source of electrons is the cathode and the electron collector is the anode. The propagating medium between cathode and anode is a plasma where the charge density ρ is much less than the density of neutral particles which are the chief source of electron scattering centers. In the plasma it is assumed that $\rho_e = \rho_i = \rho$ where ρ_e and ρ_i are charge density of electrons and ions, respectively. In analogy with photoconductors⁶ it is assumed that the equilibrium charge density in the plasma is determined by the intensity of radiation. In a qualitative manner using the concepts of

reaction kinetics,⁷ the reaction rate equation for the time-dependence of the charge density is assumed to be given by

$$\partial \rho / \partial t = k_1 R - k_2 \rho^2 - k_3 \rho, \quad (1)$$

where R is the intensity of radiation (betas, high-energy electrons, gammas, etc.), and k_1 , k_2 , and k_3 are constants independent of the charge density. The first term on the right side of the equation is the generation term, the second is a bimolecular decay term, and the third is a unimolecular decay term.

In the steady-state condition ($\partial \rho / \partial t = 0$), Eq. (1) leads to a different functional relation between ρ and R at low and high charge densities. At low charge densities, or radiation dosages, ρ should increase linearly with R and at high dosages ρ would change as R^α where α is less than unity and approaches one-half at very high dosages. These assumptions are consistent with the data obtained in reference 1 (Fig. 5 and Fig. 7) which show just this behavior under the assumption that the electron scattering in the gas is essentially independent of the radiation dosage.

In principle, once the equilibrium charge density is determined from Eq. (1), the model then requires a determination of how the average drift velocity can be evaluated as a function of voltage in the plasma. To accomplish this, a simple kinetic theory for electron transport through the plasma is outlined in which the drift velocity at the anode [$v_d(a)$ in Fig. 2] is evaluated as a function of applied anode voltage.

Consider first what may be called a very hot electron problem. The electron leaves the cathode at $x=0$ (Fig. 2) with an energy $\epsilon = \epsilon_0$. An electric field E exists in the space $0 \leq x \leq a$ as a consequence of the applied voltage V . In traversing the path between the cathode and the anode, the electron picks up a random velocity c and a directed drift velocity v_d in the x direction. The former quantity is related to the energy by the relation

$$\epsilon(x, V) = \frac{1}{2} m [c(x, V)]^2, \quad (2)$$

where m is the electronic mass. v_d is given by

$$dx/dt = v_d, \quad (3)$$

and⁸

$$v_d = (eE/m)\tau, \quad (4)$$

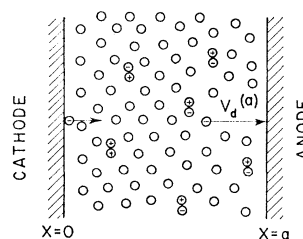


FIG. 2. Theoretical planar model.

⁷ This interpretation was suggested by Dean H. Eyring of the University of Utah.

⁸ W. Shockley, *Electrons and Holes in Semiconductors* (D. van Nostrand Company, Inc., Princeton, New Jersey, 1950), p. 200-204.

⁶ E. S. Rittner, *Photoconductivity Conference* (John Wiley & Sons, Inc., New York, 1956), p. 315.

where e is the value of the electronic charge. τ , the mean free time, is given by

$$\tau = \lambda(\epsilon)/c(x, V), \quad (5)$$

and $\lambda(\epsilon)$ is the mean free path, which is a function of energy. In traversing the interelectrode space the electron picks up energy from the electric field and loses energy by elastic collisions with the neutral scattering centers. This relation is given by⁹

$$\frac{d\epsilon}{dt} = eEv_d - \kappa\epsilon, \quad (6)$$

where κ is the fraction of the energy lost in an elastic collision event. By the use of Eqs. (2), (4), (5), and (6), the following relation:

$$\frac{1}{v_d} \frac{d\epsilon}{dt} = \frac{1}{eE} \left[(eE)^2 - \frac{2\kappa\epsilon^2}{\lambda^2} \right] \quad (7)$$

can be obtained. If the relation in Eq. (3) is then employed, the final simple differential equation is

$$\frac{d\epsilon}{dx} = \frac{1}{eE} \left[(eE)^2 - \frac{2\kappa\epsilon^2}{\lambda^2} \right], \quad (8)$$

with the boundary condition, $\epsilon = \epsilon_0$ at $x = 0$.

Before Eq. (8) can be applied to the problem, the dependence of the field E on applied voltage V must be determined. Since an ideal plasma is assumed ($\rho_i = \rho_e$), a simple solution of Poisson's equation leads to

$$E = V/a, \quad (9)$$

where a is the anode-cathode spacing. As a result, Eq. (8) becomes

$$\frac{d\epsilon}{dx} = \frac{a}{eV} \left[\left(\frac{eV}{a} \right)^2 - \frac{2\kappa\epsilon^2}{\lambda^2} \right]. \quad (10)$$

If Eq. (10) is then solved with the appropriate boundary conditions, the electron energy ϵ_a at the anode surface can be determined. By substituting this value of ϵ_a into the relation

$$v_d(a) = \frac{eV}{a(2m)^{1/2}} \frac{\lambda(\epsilon_a)}{(\epsilon_a)^{1/2}}, \quad (11)$$

one obtains the value of the drift velocity at the anode surface.

APPLICATION OF THE THEORY

The application of the theory to the case of inert gas thermionic diodes containing krypton, xenon, argon, and neon ambients is illustrated by Figs. 3-5. The boundary condition for ϵ_0 is obtained by assuming that the electrons all leave the cathode as a monoenergetic

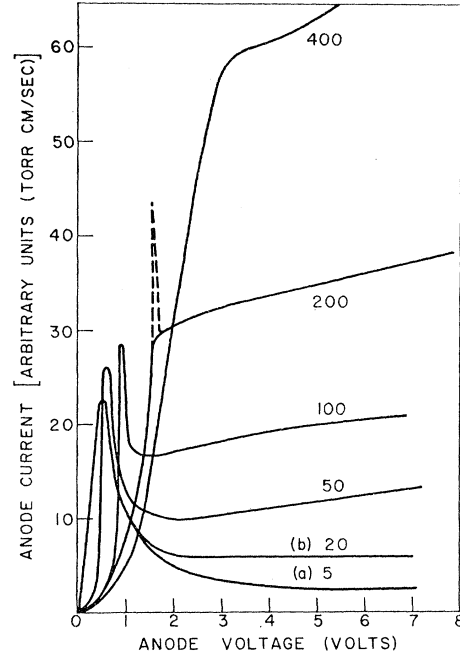


FIG. 3. Theoretical anode current-voltage characteristics for a planar thermionic diode containing krypton as the ambient gas. Cathode-anode spacing is 0.5 cm and the other parameter for each curve is the pressure in Torr.

beam in which

$$\epsilon_0 = kT_c. \quad (12)$$

In all these calculations T_c , the cathode temperature, is taken as 2000°K, under the assumption that a thoriated tungsten cathode is used and k is Boltzmann's constant. κ , the fractional energy lost per collision, is given by⁹

$$\kappa = 2m/M, \quad (13)$$

where M is the atomic mass of the element. The mean free path vs electronic energy data is obtained from the paper by Ramsauer and Kollath¹⁰ for the gases argon, krypton, and xenon, and from Normand¹¹ for neon. From the published curves, tables were compiled showing λ vs ϵ in steps of 0.1 eV over the range of interest. In interpreting between tabulated values a linear interpolation was used. The problem was treated on an LGP-30 (Act III) computer using the "SDEQ" procedure.

Figure 3 is an application of the theory to a planar diode containing krypton with an anode-cathode spacing of 0.5 cm. The ordinate for these curves is arbitrarily labeled anode current and is obtained from the calculations by the following procedure: After $v_d(a)$ is calculated at a given value of V and p , it is multiplied by the pressure (p) to obtain the value of the ordinate. This procedure assumes that the average charge density increases linearly with pressure for a constant radiation

⁹ A. von Engel, *Ionized Gases* (Clarendon Press, Oxford, 1955), p. 103-105.

¹⁰ C. Ramsauer and R. Kollath, *Ann. Physik* 3, 536 (1929).

¹¹ C. E. Normand, *Phys. Rev.* 35, 1217 (1930).

dosage. This is employed for two reasons. First, the change in drift velocity with voltage (pressure as the variable parameter) could be more graphically displayed in this way than in a plot of $v_d(a)$ vs V . In addition, the experimental data on a planar krypton diode (Fig. 6 in reference 1) indicate an increase of current, or effective charge density with increasing pressure and, to a first approximation, a linear relation is assumed.

To obtain a better understanding of the curves in Fig. 3, it is necessary to analyze Eq. (10). At very low pressures and for applied voltages in the range of ϵ_m , $(eV/a)^2 \gg 2\kappa\epsilon^2/\lambda^2$ [since $\epsilon(x) < V$]. This leads to the situation where ϵ_a is independent of pressure. Since $\lambda(\epsilon_a)$ is inversely proportional to pressure, it may be shown that $\rho v_d(a)$ is independent of pressure for the critical range of $V \cong \epsilon_m$. As a consequence, the theory predicts that in the region of maximum current all the curves for pressures below about 20 Torr merge. At high values of V and pressures below 20 Torr the value of $2\kappa\epsilon^2/\lambda^2$ approaches the value $(eV/a)^2$ which leads to a dependence of $\rho v_d(a)$ on pressure. This is illustrated in Fig. 3 which shows the merged curve at the peak, becoming two separate curves at high values of V for pressures of 20 and 5 Torr.

It should be noted that increasing the pressure above 25 Torr leads to two apparent changes: (a) The voltage at which the peak occurs moves out to higher values and (b) the width of the peak decreases. At a pressure of 200 Torr, the peak width is found to be less than 0.2 V. Since it was assumed that the electrons start from a source at 2000°K ($kT \cong 0.17$ eV), it is obvious that the detailed behavior at an apparent peak with such a small energy width would not be observed at 200 Torr. At 400 Torr, the curve has no apparent maximum.

At high pressures and low voltages, it is found that the electrons tend to lose energy to the gas in transit because $2\kappa\epsilon^2/\lambda^2$ becomes greater than $(eV/a)^2$ for $0 < x \leq a$ (e.g., in Fig. 3 this happens at a voltage of 1.5 V for the curve 200 Torr and at $V_a = 3.1$ V for 400 Torr). The more appropriate values for v_d in these circumstances are felt to be those obtained experimentally by techniques similar to those employed by Bradbury and Nielson.¹² Since this requires detailed information about v_d at E/p values of 10^{-2} V/cm Torr, which is not available for krypton or xenon,^{13,14} the curves were arbitrarily extrapolated between the origin and the first voltage value at which $d\epsilon/dx > 0$. In the case of argon and neon, where this information has been determined,¹⁵ it was used.

To sum up the analysis of Fig. 3, let us compare these theoretical curves with experimental data and determine the validity of the theory. If Fig. 3 is compared with Fig. 6 of reference 1, there is surprisingly good

agreement when one considers all the theoretical approximations used. As an example, the theory predicts that the negative resistance effect will disappear between 100–200 Torr in a krypton planar diode and the experiment shows this to happen between 90–125 Torr. The theory shows that the maximum in the curve moves to higher voltages with increasing pressure, a fact consistent with all the experimental data. Another interesting result is that the theory indicates the low-voltage part of the high-pressure curves apparently tend to shift to higher voltage values than those at a lower pressure. It should be noted that the discussion, in general, compares a planar theory with experimental results obtained using cylindrical geometry. Since the electric field near the cathode is greater than at the anode in cylindrical geometry, it can be shown using similar arguments to those used in discussing Fig. 1 that this nonuniform field would lead to an enhancement of the negative resistance effect. Under these circumstances it is felt that the application of the simple planar theory to experimental cylindrical diode experiments is valid.

The theory seems to be inconsistent with experimental data in that it shows negative resistance effects at very low pressures. However, the experimental data show that space-charge effects¹⁶ were still prevalent below 30 Torr. The theory seems to predict that if it were possible to neutralize space-charge effects, negative resistance could be observed at very low pressures. This was found to be true when measurements were made at high radiation dosage rates. Figure 8 of reference 1 shows that although negative resistance effects were not observable in a low-pressure krypton-filled (2 Torr) thermionic diode at low radiation intensities, it suddenly appeared at very high dosages. It should be pointed out here that at very low pressures (much below 1 Torr) such effects would not be measurable on a theoretical basis because the mean free path for an electron would become comparable to the cathode-anode space. Under these circumstances an electron can traverse the anode-cathode space with practically no collision (equivalent to the vacuum condition) and the proposed theory, which is based on a very large number of elastic scattering events, would not be expected to hold in this case.

The theory also predicts a much more pronounced negative resistance effect than has been observed experimentally. This behavior can be interpreted if the approximations involved in the ideal theory are considered. The theory assumes the electrons start out from a unipotential cathode as a monoenergetic beam. Both these conditions are not present in the experiment. The electrons are emitted with a Boltzmann distribution of energies and the experimental filament voltage drop is in the range of a few volts. These two practical experimental conditions tend to obscure the sharpness of the theoretical curves. In addition, inelastic collisions which

¹² N. E. Bradbury and R. A. Nielson, *Phys. Rev.* **49**, 388 (1936).

¹³ J. C. Bowe, *Phys. Rev.* **117**, 1411 (1960).

¹⁴ W. N. English and G. C. Hanna, *Can. J. Phys.* **31**, 768 (1953).

¹⁵ J. L. Pack and A. V. Phelps, *Phys. Rev.* **121**, 798 (1961).

¹⁶ R. Forman, *Phys. Rev.* **123**, 1537 (1961).

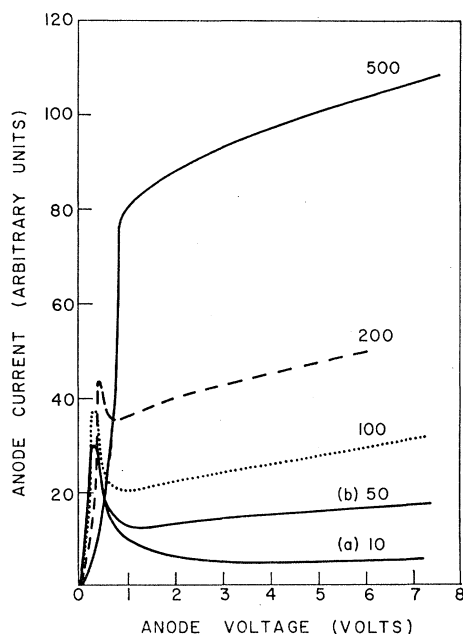


FIG. 4. Theoretical anode current-voltage characteristics for a planar thermionic diode containing argon as the ambient gas. Cathode-anode spacing is 0.5 cm and the other parameter for each curve is the pressure in Torr.

must occur experimentally should be considered. When the gas is irradiated with high energy particles not only ions but also excited states are formed. Depending on the collision cross section and density of states, scattering from these centers must become important at sufficiently high radiation dosages. The presence of inelastic scattering centers in the gas will also tend to eliminate the prominent negative effect predicted by the theory. The increase in inelastic scattering centers is probably responsible for the disappearance of experimental negative resistance effects when the radiation dosage is steadily increased at constant pressure, as seen in Fig. 7 of reference 1.

If the same type of analysis is applied to a xenon-filled planar diode, similar results, are obtained (e.g., the maximum in the curves moves to higher voltages with increasing pressure, negative resistance disappears at high pressures, etc.). In addition, a theoretical study of a xenon-filled diode shows that the negative resistance characteristic disappears at a lower pressure than that found for krypton, a result consistent with the experimental data.

Figure 4 is an application of the theory to an argon-filled diode. Again the same general features are found as have been described previously for krypton and xenon, elements all having the common feature that their electron scattering cross section has a Ramsauer minimum. The curves of Fig. 4 are different from those previously discussed for two reasons: (a) The negative resistance effect disappears at a higher pressure in argon than in xenon or krypton and (b) the maximum in the

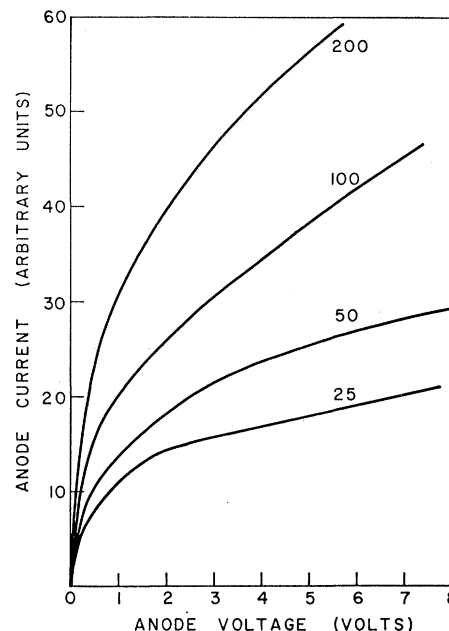


FIG. 5. Theoretical anode current-voltage characteristics for a planar thermionic diode containing neon as the ambient gas. Cathode-anode spacing is 0.5 cm and the other parameter for each curve is the pressure in Torr.

curve, especially at low pressures, appears at an extremely low voltage. The former condition is experimentally true for xenon but is not obviously so in krypton. The latter condition offers a theoretical explanation for the experimental data which show that the negative resistance effect is just detectable in argon-filled thermionic diodes and only at higher pressures. The maximum in the curves for pressures below 100 Torr occurs at 0.3 V or less. Since the thermionic source used in the experimental measurements operates at about 2000°K, it is easy to see that the details of the low-pressure curves in Fig. 4 could be masked by the inherent spread in electronic energies associated with the high-temperature emitter.

Figure 5 is conclusive evidence that negative resistance can be theoretically related to the Ramsauer minimum in the inert gases. The theoretical curves for neon in Fig. 5 show absolutely no negative resistance effects (calculations have been made up to 500 Torr) and the theoretical curves are qualitatively similar to the experimental data shown in Fig. 4 of reference 1.

In addition, the information obtained on the diatomic gas nitrogen (Fig. 9 of reference 1) is consistent with the model described in this report. Negative resistance is not observed because nitrogen does not have a Ramsauer minimum in its electron scattering cross section. In a diatomic gas the existence of inelastic collisions between electrons and nitrogen molecules at low electron energies may be assumed. The consequence of this should be: (1) lower anode currents than previously described because of lower electron drift velocities, and

(2) the absence of early breakdown effects because the electrons cannot attain very high energies in transit from cathode to anode because of inelastic collisions at low electron energies. Both of these phenomena are experimentally observed.

CONCLUSIONS

A theoretical analysis of negative resistance at low-anode voltages in inert gas thermionic diodes has led to the following conclusions. At very low pressures where the mean free path for electrons becomes comparable with anode-cathode dimensions, the current rises monotonically with increasing anode voltage. At very high pressures where the electrons tend to reach thermal equilibrium with the inert gas molecules, the current increases monotonically with increasing voltage.¹⁷ In the

¹⁷ See Reference 9, Eq. (4.21).

intermediate range of 1–300 mm, it has been shown that negative resistance can be observed, under different conditions, in gas-filled thermionic diodes containing the gases xenon, krypton, and argon which have a Ramsauer minimum in their electron scattering cross-section data. The same phenomenon does not occur in the diodes containing the gases neon or helium because they do not have this effect in their electron scattering cross section.

ACKNOWLEDGMENTS

The author would like to particularly thank Dr. H. J. Bowlden for many stimulating and critical discussions about this program. In addition, we are indebted to Dean H. Eyring, Professor M. Burton, and Dr. J. A. Ghormley for their many helpful comments during the course of this study. Finally, we would like to acknowledge the invaluable assistance of B. Kopfstein for his aid in obtaining the computer data.