$\theta_0 = 2\pi$ is about 500 W/cm^2. The measured I_0 and I_S for $\theta_0 = 0.46\pi$ are 26 and 6 W/cm^2, respectively.

[14]C. K. N. Patel and R. E. Slusher, Phys. Rev. Letters 20, 1087 (1968).

[15]R. L. Abrams and A. Dienes, Appl. Phys. Letters 14, 237 (1969).

[16]$\sigma = k/n \langle v \rangle$, where k is the slope of the linear plot and $\langle v \rangle$ the average thermal velocity of the SF_6 gas.

[17]In Ref. 14, σ was defined differently from ours by a factor of 2π. The cross section obtained in Ref. 14 is 1.2×10^{-15} cm^2.

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Ni L Self-Absorption Spectrum*†

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The shape and position of the Ni $L\alpha$ x-ray emission line ($\lambda = 14.53$ Å) has been studied as a function of bombarding electron energy ranging from 2 to 30 keV. The bombarding electrons struck the anode at normal incidence, and x rays were viewed by the spectrometer at a 90° takeoff angle. The Ni $L\alpha$ lines recorded under these conditions exhibit an increasing attenuation of intensity on the high-energy side and a shifting of the peak position towards lower energy with increasing bombarding electron energy. These changes are interpreted as due to differential self-absorption of the emission line in the anode. The emission measurements have been used to obtain the "L self-absorption spectrum" of Ni, which agrees favorably with the absorption spectrum reported in the literature.

INTRODUCTION

The Ni $L\alpha$ x-ray emission line represents the transitions of valence band ($M_{IV,V}$) electrons to vacancies in the L_{III} shells of Ni atoms. It is believed by some authors that if the combined width of the Ni L_{III} states and the spectral-window function of the instrument which records the spectrum is small compared with the width of the Ni valence band, one can learn from the shape of the observed Ni $L\alpha$ line the density of filled valence states in Ni. For purposes of gaining such information, various authors[1-11] have recorded the Ni $L\alpha$ line. It is a significant fact that almost no agreement exists regarding the detailed features of the line and of the location of its peak position. The differing experimental results and the attempted explanations of various authors suggested that a systematic experimental investigation of the Ni $L\alpha$ emission line as a function of bombarding electron energy ranging from 2 to 30 keV might clarify the unsettled points in the interpretation of x-ray spectra. As a result of the present study, it has been found that the changes in the shape and position of the Ni $L\alpha$ line are related to the self-absorption of radiation by the material of the anode.

EXPERIMENTAL

The data on the Ni $L\alpha$ x-ray emission line spectra were recorded with the two-crystal vacuum x-ray spectrometer at New Mexico State University. Potassium acid phthalate (KAP) crystals ($2d = 26.6$ kxu) were used as diffracting elements, which were grown from a supersaturated water solution in this laboratory. The excellent spectrometric properties of KAP crystals for the wavelength range of the present investigation have already been established.[12]

The spectral window, implied by the width of the (1, −1) curve at the Ni $L\alpha$ wavelength, is $\simeq 0.6$ eV (resolving power $\lambda/d\lambda \sim 3000$), which is good enough to reproduce satisfactorily the structure on the high-energy side of the Ni $L\alpha$ line previously reported. The data were obtained by the operation of the two-crystal spectrometer in a double-rotation tracking mode.[13] The term "double" implies that both crystals are rotated for scanning the spectrum, as compared to the rotation of the first (central) crystal only in the case of the single-rotation mode. In the method of double rotation, x rays which pass through the instrument originate from the same region of the anode, strike the same two areas of the crystals, and enter the detector at the same position for every setting of the spectrometer. Thus, distortions resulting from possible nonuniformities of the source or inhomogeneities of the crystals are eliminated. The effects of beam walking were noted in the tail regions of the Ni $L\alpha$ line where the intensity in the single-rotation mode falls below that obtained in the double-rotation mode.

The detection system used in the investigation consisted of a flowing-gas (P-10) proportional counter followed by a linear amplifier, a single-channel pulse-height analyzer, and an electronic scaler and timer.

The target self-absorption plays a significant role in determining the spectrum of valence-band electron to inner-shell hole transitions in metals. This effect is crucial for the Ni L emission spectrum where the absorption coefficient in the vicinity of the L_{III} edge undergoes large fluctuations because of the relatively narrow and partly empty $3d$ band.[14] For this reason, one of the major concerns in designing the experiment was the explicit determination of the effects of target self-absorption. This was accomplished by employing normal incidence for the bombarding electrons and approximately a 90° takeoff angle for the x rays. A demountable continuously pumped x-ray tube was used. The condensation of pump oil vapors and filament evaporants on the surface of the anode produces serious target contamination. To avoid this, the filaments were screened from the anode by water-cooled semicylindrical shields to avoid direct contamination of the anode by filament evaporants. At low bombarding electron energies ($\simeq$ 2 keV) the contamination of the anode proved to be disastrous, even at such low operating pressures as 1×10^{-7} Torr. Since the counting rates were quite low for low bombarding electron energies, the additional intensity reduction due to target contamination prevented successful experiments. An attempt was made to avoid this contamination by the use of a hot anode. The x-ray tube was operated so that the thin sheet (20 mil thick) of electropolished grade A nickel appeared red hot ($T \simeq 800$°C). However, the target was not so hot that the x-ray tube walls showed any noticeable traces of evaporated Ni. The hot target was used successfully as a stable source of radiation for both low and high bombarding electron energies. In order to transmit as much radiation as possible, thin vinyl plastic[15] windows with an x-ray transmission of about 80% at the Ni $L\alpha$ wavelength were used for the x-ray tube and the detector. Separate voltage regulated power supplies were employed for the x-ray tube and detector.

RESULTS

The results of this systematic study of the Ni $L\alpha$ line shape and position as a function of bombarding electron energy are presented in Fig. 1. The line was recorded at different bombarding electron energies in the range of 2 to 15 keV. The ordinate values represent the intensity of the Ni $L\alpha$ radiation which was determined to 1% or better at the line peak in all the measurements. The background counting rate for the proportional counter was 10 counts per min. The data have been normalized to the same peak intensity. The zero of energy is arbitrarily chosen to be at the peak of the 2-keV curve. Since the shape of the line was the prime interest of this investigation, no wavelength calibration was attempted.

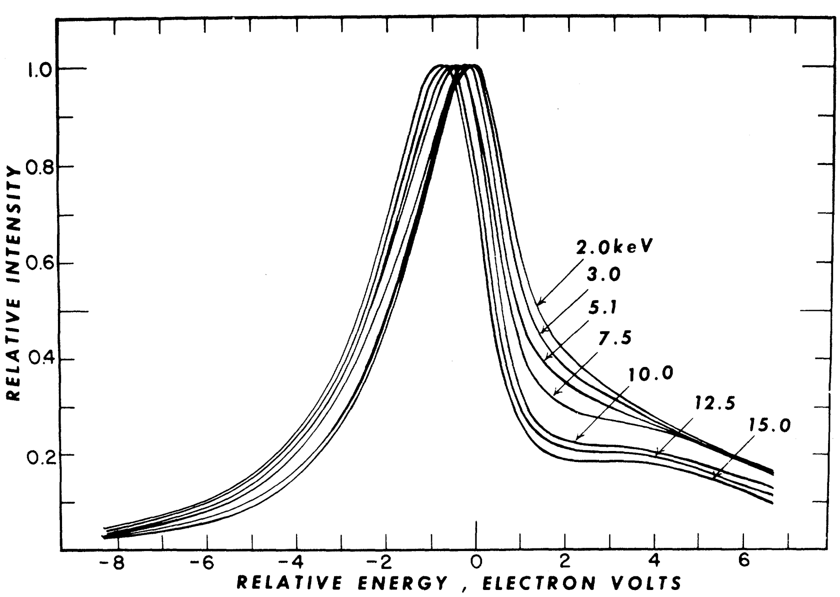

FIG. 1. Ni $L\alpha$ line profiles for several bombarding electron energies.

It is evident that the profile of the Ni $L\alpha$ line changes radically as a function of bombarding electron energy. The emission spectrum shows an increasing attenuation of the intensity on the high-energy side of the line and a shifting of the peak position towards lower energy. The shift is approximately 0.7 eV in the case of the 15-keV peak relative to the 2-keV peak. When the bombarding electron energy is of the order of 5 keV, the ramp on the high-energy side of the line takes on the appearance of a low foot. This structure is much like that previously reported by Spielberg, Soules, and Shaw[9] and the single-crystal work of van den Berg.[11] The foot is preceded by a relative minimum of intensity located at approximately 1.6 eV from the 2-keV line peak. These features gain prominence at higher bombarding electron energies.

DISCUSSION OF EXPERIMENTAL RESULTS

Of most direct interest for the purposes of the present discussion are: (i) an increasing attenuation of intensity on the high-energy side of the line, (ii) a shifting of the line position towards lower energy, and (iii) the gaining in prominence of the low foot on the high-energy side of the line with increasing bombarding electron energy. The changes in the shape of the Ni $L\alpha$ line must be related to some phenomenon whose ability to reduce the intensity on the high-energy side of the line increases with increasing bombarding electron energy. The electrons penetrate deeper into the material at high bombarding electron energies and the effective depth of production of the Ni $L\alpha$ photons increases. In the case of Ni, the absorption transitions involving the excitation of L_{III} electrons to a relatively large density of empty $3d$ states just above the Fermi energy cause the coefficient of absorption μ to undergo a large step, generally characterized by a "white" absorption "line." Thus, if the Ni $L\alpha$ radiation traverses a significant amount of the anode material before emerging, the large changes in μ could effect a noticeable change in the intensity distribution on the high-energy side of the line. Excitation or satellite bands located on the high-energy side of the parent Ni $L\alpha$ line are also subject to this attenuation.

In the source geometry which we used, the effective depth of production of Ni $L\alpha$ radiation varies with the bombarding electron energy. At 15 keV the effective depth is much greater than that at 5 keV.[16] The 15-keV curve should, therefore, show considerably less intensity on the high-energy side of the line than the 5-keV curve if self-absorption of the radiation by the emitting anode plays an important role. Such a difference is evident in Fig. 1. The curves recorded at intermediate bombarding electron energies fit the above picture very well, and substantiate the quantitative conclusions relating these changes of intensity to the self-absorption of Ni $L\alpha$ radiation by the emitting anode.

The attenuation of the spectrum on the high-energy side of the Ni $L\alpha$ line produces an exaggerated asymmetry which shifts the energy position of the line peak towards lower energy. This shift will vary according to the degree of attenuation of intensity on the high-energy side of the Ni $L\alpha$ line, and will therefore increase at higher bombarding electron energies. This interpretation is completely borne out by the present investigation. Some authors[17,18] have attempted to connect these asymmetries with the band structure of the solid. According to their interpretation, the decrease of intensity on the high-energy side of the emission line occurs because beyond a certain point the bands are no longer filled, and the sharpness of the decrease is limited only by the width of the inner state. They rule out the possibility that anode self-absorption of the high-energy side of the emission line had any part in producing the high asymmetries, although their curves were recorded with small takeoff angle and with bombarding electron energies as high as 35 keV. Hanson and Herrera[19] have demonstrated that self-absorption probably did modify the $K\beta_5$ curves of Bearden and Shaw.[17] In the present experiments, a bombarding electron energy of 30 keV produced sufficient self-absorption to reduce the intensity of the high-energy side of the Ni $L\alpha$ line to a vanishingly small amount. Some investigators,[20] who have inadvertently obtained similar results, have mistakenly interpreted the chopped-off emission line as showing a good Fermi edge. In the literature,[21] a few instances of apparent coincidence of such an emission edge with the corresponding absorption edge in metals have been reported. The coincidence may not exist after proper corrections for the effects of self-absorption are made.

The low foot or small hump on the high-energy side of the Ni $L\alpha$ line has been interpreted as a satellite of the parent line by some investigators.[5,8,10] In the present investigation this hump appears at bombarding electron energies of about 5 keV and becomes quite prominent at higher bombarding electron energies. This feature is satisfactorily explained in terms of the self-absorption of the emission line shape. Figure 2 shows the Ni $L\alpha$ lines recorded at 2 and 15 keV. The L_{III} absorption spectrum of Ni according to van den Berg[11] has also been included. All the curves have been plotted on the same energy scale. It is evident that the observed minimum on the high-energy side of the relatively more self-absorbed curve, i.e., the 15-keV curve, coincides with the peak of the white absorption line of the L_{III} absorption spectrum. The intensity of the emission curve goes up again towards higher energy because of the considerable decrease in the absorption coefficient μ in that energy region and finally the intensity

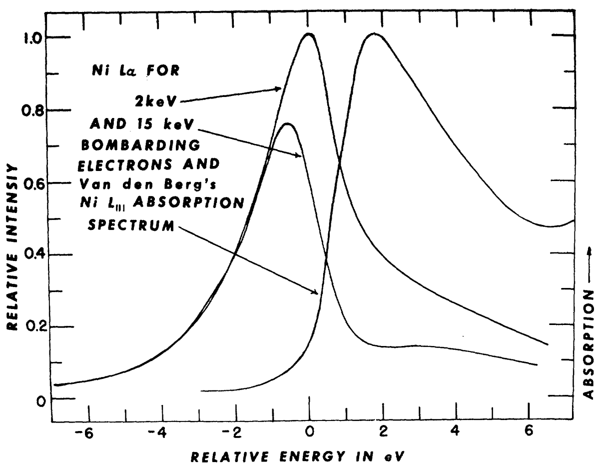

FIG. 2. Apparent coincidence of the observed minimum on the high-energy side of Ni $L\alpha$ line recorded for 15-keV bombarding electrons with the maximum of L_{III} absorption spectrum of Ni.

levels off. The low foot becomes more prominent with the increase in differential absorption, implied by the greater path length for higher-energy electrons. The low foot is, therefore, interpreted to be a result of the differential anode self-absorption of the satellite emission which exists on the high-energy side of the Ni $L\alpha$ line and is not a new satellite of the parent Ni $L\alpha$ line.

On the low-energy side of the Ni $L\alpha$ line, the absorption coefficient is low and does not show any appreciable abrupt variations. Therefore, any significant change in the shape on the low-energy side of the Ni $L\alpha$ line is not expected. This fact is amply supported by the present investigation.

If our interpretation that the changes in shape and position of the Ni $L\alpha$ line are connected with anode self-absorption is correct, then a self-absorbed Ni $L\alpha$ emission line could be considered as resulting from the attenuating influence of the L_{III} absorption spectrum on the basic Ni $L\alpha$ line. The attenuation of the intensity is described by the equation $I/I_0 = e^{-\mu x}$, where I_0 is the original intensity of the beam and I is the intensity after traversing a thickness x of absorbing material whose absorption coefficient is μ. Therefore, a plot of the ratios of the intensities of the two curves – one grossly self-absorbed (e.g., for $E_{inc} = 16$ keV) and the other having negligible self-absorption (e.g., for $E_{inc} = 3$ keV) – should reflect a quantitative picture of the L_{III} absorption spectrum of Ni. Figure 3 shows the results of such a calculation where the measurements have been extended to include the emission spectra of Ni in the $L\alpha$-$L\beta$ region. The line intensities of the two emission spectra have been normalized to match on the low-energy side of the $L\alpha$ line. It is noted that the computed curve has striking similarities with the L photon absorption spectrum of Ni. Both the L_{II} and L_{III} absorption edges stand out clearly and all the fine-structure bumps reported earlier show nicely also. This point-by-point plot of the ratio of the intensities of two emission lines recorded with 3- and 16-keV bombarding electrons, respectively, is termed here as "self-absorption spectrum." On the whole, the agreement is remarkably good and confirms the interpretation that the increasing attenuation of the line intensity on the high-energy side, the shifting of the line position towards lower energy, and the appearance of the low foot on the high-energy side with increasing bombarding electron energy are due to the self-absorption of the x rays by the material of the emitting anode.

The relative positions of any two points on a computed curve of $\ln[I(16\text{ keV})/I(3\text{ keV})]$ versus energy determine $(\mu_1 - \mu_2)x_{eff}$, where x_{eff} is defined as the mean depth at which the entire luminosity could be concentrated without change in the external observed intensity. The difference in the values of the absorption coefficients, $\mu_1 - \mu_2$, corresponding to these points can be found from the L_{III} photon absorption coefficients as reported by Cauchois and Bonnelle.[22] Thus, in principle, an estimate of the effective depth of the production of L_{III} holes in Ni at any bombarding electron energy in excess of 3 keV can be obtained. For accurate determination of the effective depth, both emission spectra should be corrected for finite resolving power of the instrument, scattered background radiation, and nonlinear response of the detector. However, for recording with a two-crystal spectrometer equipped with KAP crystals (resolving power $\simeq 3000$) and photon-counting system, these corrections are usually small. Further, the photon absorption spectrum of Ni, to be used in conjunction with the present self-absorption spectrum, should also be subjected to similar corrections. The photon absorption spectrum mentioned above was recorded with a curved crystal spectrograph and not corrected for

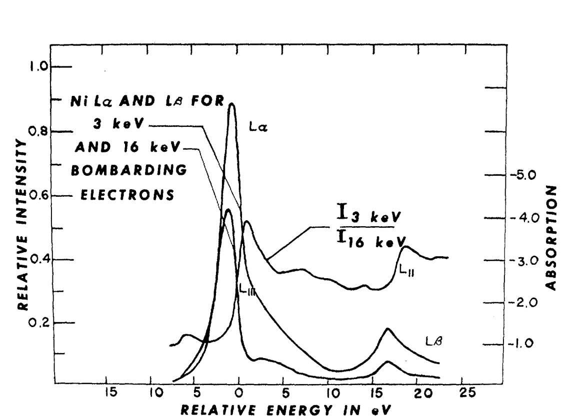

FIG. 3. Ni L self-absorption spectrum obtained from the ratio of the intensities of the emission spectra in the Ni $L\alpha$-$L\beta$ region for 3- and 16-keV bombarding electrons.

the instrumental effect. The above corrections are, therefore, not attempted, and the results of the calculation of the effective depth of production of L_{III} holes in Ni are semiquantitative. Numerical calculations were made for the values of the effective depth of the production of L_{III} holes in Ni according to Makhov's method.[23] These values were found close to the present experimental values in the region of low energies. Therefore, the effective depth at energy 3 keV from Makhov's method was added to the experimental result. Figure 4 presents experimental values of the effective depth of the production of L_{III} holes in Ni computed in this manner. The corresponding values calculated on the basis of existing semiempirical electron retardation relations[23, 24] are also included for comparison purposes. It should be noted that the values of effective depth of the production of x rays used by other investigators for assessing the effects of anode self-absorption were probably too high.

It is important to note that this investigation suggests that it is possible to obtain an absorption spectrum of Ni from valence-band emission line shapes recorded at two widely different bombarding electron energies. The technique is advantageous for studying the absorption and emission spectra of the same material in bulk form, and it eliminates the tedious process of preparing ultrathin absorbers.

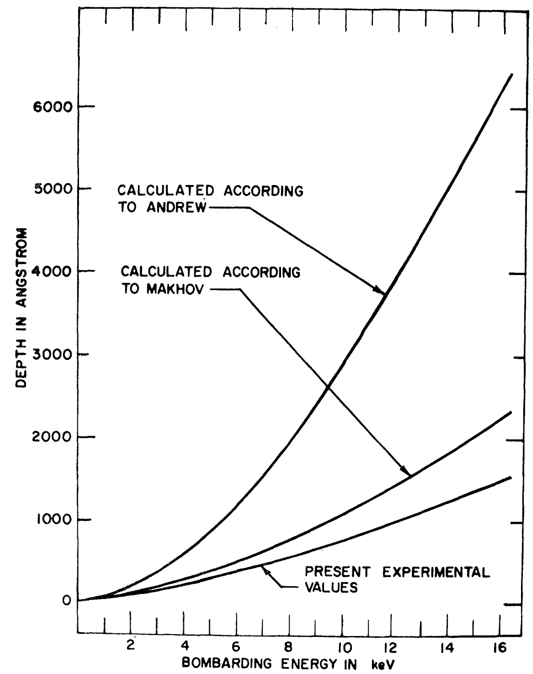

FIG. 4. Comparison of present results regarding the effective depth of production of L_{III} holes in Ni with the results of other authors.

CONCLUSIONS

For electron energy increasing from 2 keV, the Ni $L\alpha$ emission line exhibits an increasing attenuation of intensity on the high-energy side and a shifting of the line position towards lower energy. These changes have been considered in terms of the differential self-absorption of the radiation in the anode. It has been shown that the recorded Ni $L\alpha$ curves fit the above interpretation in a satisfactory fashion. It is suggested that it is possible to record the Ni $L\alpha$ emission line with a negligible amount of anode self-absorption by using sufficiently low bombarding electron energies and a suitable x-ray source geometry. Of all the valence-band spectra recorded to date it is significant to note that few, if any, are free of the distorting effects of anode self-absorption for reasons of both excessive bombarding electron energies and unfavorable anode geometries. Emission-line measurements have been used to obtain a quantitative picture of the L "self-absorption spectrum" of Ni. These data, in conjunction with the reported data on photon absorption coefficients of Ni, have been used to evaluate the effective depth of production of L_{III} holes in Ni for bombarding electrons of several energies.

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†For preliminary discussion of this work, see D. Chopra and R. Liefeld, Bull. Am. Phys. Soc. 9, 404 (1964).

[1] R. Thoraues, Phil. Mag. 2, 1007 (1926).

[2] J. Shearer, Phil Mag. 20, 504 (1935).

[3] F. C. Chalklin, Nature 135, 998 (1935); Proc. Roy. Soc. (London) A155, 366 (1936).

[4] F. Tyren, Z. Physik 98, 768 (1938).

[5] J. Farineau, Nature 140, 508 (1937); Ann. Phys. (Paris) 10, 20 (1939).

[6] E. Gwinner, Z. Physik 108, 523 (1938).

[7] H. W. B. Skinner, Rept. Progr. Phys. 5, 257 (1939).

[8] Y. Cauchois, Phil. Mag. 44, 173 (1953).

[9] N. Spielberg, J. A. Soules, and C. H. Shaw, Tech. Rept., Contract No. N5ONR-22521, Office of Naval Research, Washington, D. C.

[10] H. W. B. Skinner, T. G. Bullen, and J. E. Johnston, Phil. Mag. 45, 1070 (1954).

[11]C. B. van den Berg, Ph.D. thesis, Groningen University, 1957 (unpublished).
[12]R. Liefeld, D. Chopra, W. Gray, and D. Mott, Bull. Am. Phys. Soc. 8, 313 (1963).
[13]S. K. Allison, Phys. Rev. 44, 63 (1933).
[14]See L_{III} photon absorption spectrum of Ni (Ref. 11).
[15]Vyns-3 Blend B-1494 Union Carbide Plastics Co., N.Y.
[16]C. Feldman, Phys. Rev. 117, 455 (1960).
[17]J. A. Bearden and C. H. Shaw, Phys. Rev. 48, 18 (1935).
[18]W. W. Beeman and H. Friedman, Phys. Rev. 56, 392 (1939).
[19]H. P. Hanson and J. Herrera, Phys. Rev. 105, 1483 (1957).
[20]H. W. B. Skinner, Rept. Progr. Phys. 5, 257 (1939).
[21]See, for example, W. M. Cady and D. H. Tomboulian, Phys. Rev. 59, 381 (1941).
[22]Y. Cauchois and C. Bonnelle, Compt. Rend. 245, 1230 (1957).
[23]A. F. Makhov, Fiz. Tverd. Tela 2, 2161 (1961) [English transl.: Soviet Phys. – Solid State 2, 1934 (1961)].
[24]V. J. Andrew, Phys. Rev. 42, 591 (1932).

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Theory of the Forward Peak in the Angular Distribution of Electrons Ejected by Fast Protons*

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The cross section for ejecting electrons by 300-keV protons is calculated using the first term in the Neuman expansion of Faddeev's equation for the final state of the electron-proton-residual ion system. This approximation predicts a peak at 0° in the angular distribution of electrons ejected with a velocity approximately equal to the velocity of outgoing protons. Numerical results for He and H_2 target gases are given and compared with the experiments of Rudd and co-workers. The qualitative behavior of the forward peak in the experimental angular distributions is well accounted for.

I. INTRODUCTION

Experimental techniques have reached the stage where the energy and angular distribution of electrons ejected by heavy ion impact can be measured. Because cross sections that are differential in both the energy and angle of the ejected electron are obtained, these experiments produce much valuable information about the ionization process. In particular, Rudd, elaborating on a suggestion by Oldham,[1] has proposed a new mechanism for ionization to explain the forward peaking in the published data[2] on the ionization of He and H_2 by proton impact. Since the forward peaking is most pronounced when the electron is ejected with a velocity nearly equal to the velocity of the proton, Rudd argues that some electrons are carried along with the proton for a short time and then move away from the proton as free electrons. This paper discusses the theoretical basis for Rudd's mechanism, and shows in just what sense an electron in continuum state is carried along by a proton.

We seek a first-order approximation for the final-state wave function of the electron, proton, and residual ion which can describe an electron being carried along by the proton. Of equal importance is the requirement that the approximate wave function give a reasonably good indication of what a more accurate theory would predict. The first term in the Neumann expansion of Faddeev's[3] equation for the final-state wave function exactly satisfies these two requirements. Since our approximation is only first order, its range of validity is considerably restricted at the outset. The highest energy for the proton in the experiments of Rudd *et al.* was 300 keV. Since a 300-keV proton is only moving with a velocity of about 3.5 times the velocity of an electron in the first Bohr orbit of the hydrogen atom, a first-order approximation generally does not adequately describe the detailed distribution of electrons in energy and angle. A plane wave accurately describes the motion of the proton, but the wave function of the ejected electron is distorted by both the field of the residual ion and the field of the proton. Since