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## Observation of Beam-Induced Changes in the Polarization of Balmer- $\alpha$ Radiation Emitted Following Beam-Tilted-Foil Transmission

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## Observation of beam-induced changes in the polarization of Balmer- $\alpha$ radiation emitted following beam-tilted-foil transmission

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Measurements of the circular polarization of Balmer- $\alpha$  radiation emitted by excited hydrogen atoms, following the transmission of (20–50)-keV protons through thin, tilted amorphous carbon foils, exhibit markedly unexpected behavior as a function of exposure of the foil to the proton beam. Specifically, the circular polarization changes from an initially well understood tilt-angle dependence to a behavior which, for low tilt angles, gives the *opposite handedness of circular polarization* from that predicted. In addition, the degree of alignment, indicated by the linear Stokes parameter  $M/I$ , is enhanced also as a function of dose. These changes in the tilt-angle dependence of the Stokes parameters have been systematically correlated with beam-induced graphitization of the foil, which is observed to occur from Raman measurements.

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### I. INTRODUCTION

The elucidation of the nature of electron exchange which occurs between an atomic system and a nearby surface is a fundamental issue in surface science. Indeed, a comprehensive understanding of this basic particle-surface interaction does not presently exist. One way to investigate this electron-exchange process is to monitor the final states of excited atoms created by transmission of ions through a thin foil. Measurements of both the intensity and polarization of the radiation emitted as these atoms decay to lower energy levels provide a fingerprint of the state of the atom at the instant of electron capture and can provide clues into the mechanisms involved in the electron-exchange process itself.

The transmission of an energetic beam of ions through a very thin foil is an extremely sudden event which results in the neutralization and coherent excitation of many of the ions in the beam. During passage of the ion through the foil, an ongoing electron exchange is possible where atomic states of the projectile are occupied by resonant capture of electrons from the foil and later depopulated by resonant ionization. The final state is determined after the projectile is some distance away from the surface, often called the freezing distance, where such capture-loss processes are no longer possible. Due to the high degree of spatial anisotropy inherent in this impulsive excitation of the beam particles by transmission through the foil, the excited states often possess a non-uniform population of magnetic sublevels. This fact can be understood by noting that the Galilean transformation of the foil band structure—necessary because of the relative motion of the projectile and foil—brings only a subset of the filled conduction-band states into resonance with the atomic levels of the projectile. Therefore the foil electrons which are capable of participating in the electron exchange have a pre-

ferred direction in momentum space [1]. These electrons will then be captured into select angular momentum substates of the excited species. Thus the magnetic-sublevel electron distribution of the ensemble of beam-foil-excited states is manifest in the polarization of the radiation emitted as these states decay to lower energy levels; and the analysis of such polarization can provide insight into the state of the atom at the instant of electron capture and the dynamics of the electron-capture processes which lead to the neutralization and excitation of the transmitted beam.

In this paper we report on recent measurements of the polarization of Balmer- $\alpha$  radiation emitted by excited hydrogen atoms following the transmission of (20–50)-keV protons through thin, tilted, initially amorphous carbon (*a*-C) foils. These polarization measurements show changes which occur as a function of the total integrated beam dose. The observed changes in the polarization of the beam-foil radiation have been correlated to an ion-beam-induced change in the structure of the foil. Raman measurements of the foil samples before and after irradiation by the proton beam indicate that proton bombardment modifies the original amorphous structure of the foil into a structure which is more graphitic in character. Thus the proton beam itself, which is used to probe the electron exchange occurring at the foil surface, modifies the foil structure and influences the outcome of the electron-exchange interaction. The tilt-angle dependence of the polarization of the Balmer- $\alpha$  radiation emitted by the foil-excited hydrogen atoms also changes with proton dose. Of particular interest is the drastic and unexpected change in the tilt-angle dependence of the circular polarization. Initially the circular polarization is nearly a linear function of the tilt angle, being zero for normal incidence, and predominantly right-hand (left-hand) circular for positive (negative) tilt angles. This type of tilt-angle depen-

dence of the handedness of the circular polarization is expected and is easily understood from simple symmetry arguments. However, after the foil graphitization, which is caused by prolonged proton irradiation, the circular polarization is no longer a monotonic function of the tilt angle. Rather the tilt-angle dependence of the circular polarization changes to an oscillatory behavior which, for small tilt angles, has the *opposite* handedness to the initial, well-understood, expected case.

Our measurements clearly show that changes in the polarization of the emitted Balmer- $\alpha$  radiation are correlated with changes in structure of the foil, which in turn show a very strong dependence on the total integrated beam dose. It is important to note that the changes in foil structure and in the Balmer- $\alpha$  polarization can be brought on more quickly by irradiating at larger current densities; however, they still occur when one irradiates the foil at lower current densities for a longer period of time. This observation is substantially different from that of Hight *et al.* [2] and Gay and co-workers [3,4] where they report on normal-incidence alignment measurements of He I radiation following transmission of (40–1300)-keV helium ions through  $\alpha$ -C foils. They found that the alignment increased with beam-current density and this increase was correlated to an increase in the foil temperature and an associated decrease in the secondary-electron yield. They proposed a model where the secondary electrons produce random microscopic fields which tend to reduce the alignment. The rate of secondary-electron production per incident ion is larger at small beam-current densities and as a result a smaller alignment is observed. Winter [5] observed a similar current dependence in the normal-incidence alignment of He I radiation. In contrast, the measurements we present here using protons in the (20–50)-keV energy range show an increase in the Balmer- $\alpha$  alignment at normal incidence and unexpected changes in the orientation as a function of tilt angle, which each vary with the *total accumulated beam dose*, rather than with the *instantaneous beam-current density*.

The different systems studied and beam energies used in our work compared to the works of Hight *et al.*, [2] Gay and co-workers [3,4], and Winter [5] makes comparison of the results somewhat difficult. However, one additional difference that should be pointed out is the use of a nickel mesh to support our carbon foils. Due to the lower beam energies and long analysis times used in our experiments, the foil had to be mounted on a 90% transmission nickel mesh in order to survive for a long enough period of time to perform the experiment. The differences in the beam-fluence versus beam-current dependence of the polarization of radiation emitted by the beam-foil-excited states in our experiments compared to Hight *et al.*, [2] Gay and co-workers [3,4], and Winter [5] may be attributed to an unknown influence from the nickel mesh. However, our very early experiments without a nickel mesh do not suggest this to be the case.

## II. EXPERIMENTAL METHODOLOGY

The experiments described herein were performed in an ultra high-vacuum chamber with a base pressure of  $2 \times 10^{-10}$  Torr. A beam of protons, produced by a Colutron ion source, focused by an einzel lens, and mass analyzed by

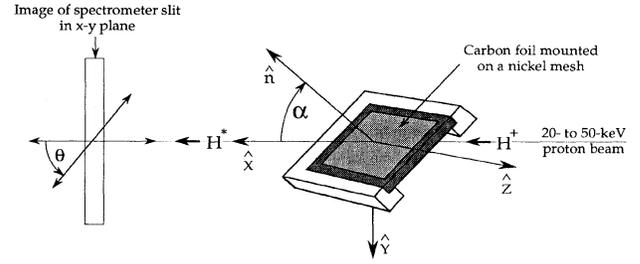


FIG. 1. The beam-tilted-foil sample geometry is shown. A (20–50)-keV proton beam passes through a thin carbon foil. The normal to the foil makes an angle  $\alpha$  with the beam. Following transmission through the foil, the proton captures an electron into an excited state. The intensity and polarization of the deexcitation radiation emitted by the hydrogen atom is measured downstream from the foil.

a Wien filter, was incident on a very thin carbon foil arranged in the UHV chamber in the transmission geometry. A significant fraction (on the order of one-half) of the protons captured an electron upon passage and emergence from the foil, creating neutral, sometimes excited, hydrogenic states. The intensity and polarization of the Balmer- $\alpha$  radiation emitted by the foil-excited hydrogen atoms were monitored as a means of characterizing the electron-exchange interaction.

The experimental geometry of the foil target is depicted in Fig. 1. The samples used for these experiments were amorphous carbon foils ranging in surface density from 0.5 to 2.0  $\mu\text{g}/\text{cm}^2$ . The foils were mounted onto a  $2 \times 6$  cm<sup>2</sup> rectangular frame which had been covered with a 90% transmission, 70 wire per inch, nickel mesh. The mesh was absolutely necessary for support due to the extreme thinness of the foil. The energetic ion beam, incident along the  $\hat{x}$  axis, impacted the foil sample on the nickel mesh side and emerged on the mesh-free side. The foil could be rotated about the  $\hat{z}$  axis and the tilt angle  $\alpha$ , measured from the  $\hat{x}$  axis to the surface normal, described the degree of rotation. The sense of rotation was such that a rotation of the foil which moved the surface normal toward the positive  $\hat{y}$  axis represented a positive tilt angle. For example, the orientation of the foil depicted in Fig. 1 is described by a negative value of  $\alpha$ . Rotation of the foil could be accomplished either manually or under computer control.

The optical radiation emitted by the foil-excited atoms was first collimated by a lens inside the vacuum chamber before passing through a polarimeter. The polarimeter, based on a design described by Berry, Gabrielse, and Livingston [6], consisted of a rotatable zero-order quartz retardation plate followed by a fixed linear polaroid. The phase delay of the retarder was measured to be  $0.253 \pm 0.002$  of a full wavelength at 6563 Å. Light exiting the polarimeter was spectrally analyzed with a monochromator and cooled photomultiplier tube operating in single pulse counting mode. Measurement of the photon intensity as a function of angular position of the quarter-wave plate allowed one to determine the reduced Stokes parameters

$$\frac{M}{I} = \frac{I_{0^\circ} - I_{90^\circ}}{I_{0^\circ} + I_{90^\circ}}, \quad \frac{C}{I} = \frac{I_{45^\circ} - I_{-45^\circ}}{I_{45^\circ} + I_{-45^\circ}}, \quad \frac{S}{I} = \frac{I_{\text{RCP}} - I_{\text{LCP}}}{I_{\text{RCP}} + I_{\text{LCP}}}, \quad (1)$$

which describe the polarization state of the emitted radiation [6,7]. In the above definitions,  $I_\theta$  is the intensity of light linearly polarized at  $\theta^\circ$  with respect to the  $\hat{x}$  axis (see Fig. 1), and  $I_{\text{RCP}}$  and  $I_{\text{LCP}}$  are the intensities of right- and left-hand circular polarized light, respectively.

The data presented in this paper were acquired by rotating the foil to a desired tilt angle with a computer-controlled stepper motor and then measuring the intensity and the polarization (Stokes parameters) of the Balmer- $\alpha$  radiation emitted by the beam-foil-excited states. Typical tilt-angle spectra were obtained by measuring the Stokes parameters at discrete tilt-angle positions between  $\alpha = -75^\circ$  and  $+75^\circ$ , usually starting at  $\alpha = 0^\circ$  and working symmetrically outward toward the larger tilt angles (i.e., a typical sequence was  $\alpha = 0^\circ, 6^\circ, -6^\circ, \dots, 72^\circ, -72^\circ$ ). Tilt angles larger than  $\pm 75^\circ$  were not feasible because the count rate was reduced due to the blocking of the beam by the nickel mesh. At each setting of the foil tilt angle a measurement of the intensity  $I$  and the reduced Stokes parameters  $M/I$ ,  $C/I$ , and  $S/I$  required between one and four minutes depending upon the photon count rate and the desired precision of the measurement. Stability of the proton beam was generally sufficient to allow uninterrupted data acquisition for several hours at a time. In order to ensure that the measurement was unaffected by any beam instabilities, missed steps by the polarimeter, or any other problems, the reduced chi square ( $\chi^2_\nu$ ) for the fit was computed for each polarization measurement. If a high value of  $\chi^2_\nu$  was encountered then the origin of the problem was investigated and corrected and the measurement was repeated.

Using the procedure described above, we made measurements of the tilt-angle dependence of the Stokes parameters of Balmer- $\alpha$  radiation emitted by beam-foil-excited hydrogen atoms. These measurements, described in Sec. IV, were found to depend on the preirradiation history of the carbon foils.

In order to determine the effect of ion-beam irradiation on the carbon foil samples, we made comparisons of the Raman spectra of foils before and after irradiation by the proton beam. The Raman measurements, made at Brigham Young University, utilized an argon-ion laser operating at a wavelength of 4880 Å. The laser beam was incident on the foil sample at  $70^\circ$  from the surface normal and its power was approximately 100 mW at the foil surface. Measurements of the Raman spectra were made using a SPEX 1877 Triplemate Raman spectrometer. The results of these measurements are presented in Sec. III.

### III. RAMAN SPECTRA OF UNIRRADIATED AND IRRADIATED EVAPORATED $a$ -C FOILS

As will be shown in Sec. IV, measurements of the tilt-angle dependence of the polarization of Balmer- $\alpha$  radiation emitted by foil-excited hydrogen atoms exhibit a strong dependence on the cumulative exposure of the foil to the incident proton beam. The interpretation of these beam-dose-dependent data requires an understanding of the structural metamorphosis afforded to the carbon foil by the interaction of the proton beam with the atoms in the foil. This section will describe these ion-beam-induced changes which affect

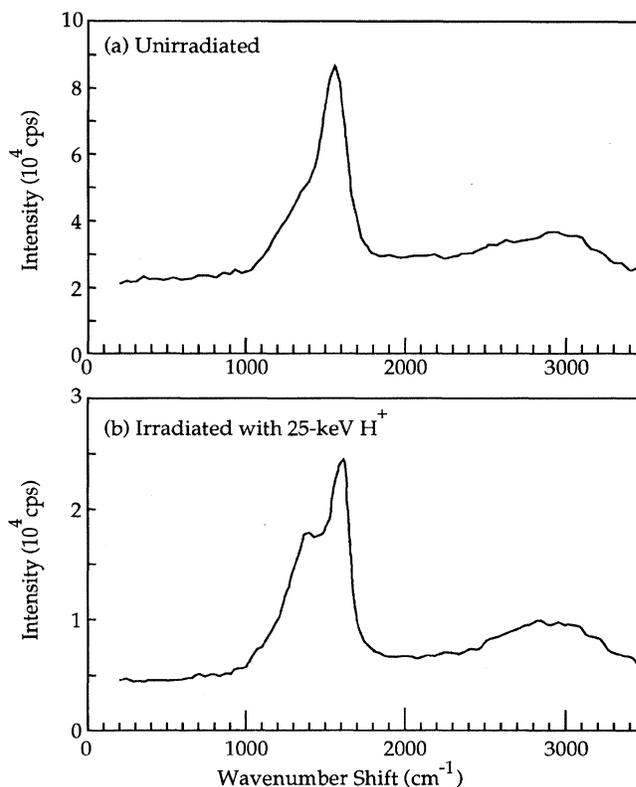


FIG. 2. Raman spectra of a  $2.0\text{-}\mu\text{g}/\text{cm}^2$   $a$ -C foil are shown. The spectrum is from (a) an unirradiated spot and (b) a spot which had been irradiated with a  $0.5\text{-}\mu\text{A}/\text{mm}^2$ , 25-keV proton beam to a total dose of approximately 50 mC.

the structure and bonding of atoms in the irradiated portion of the carbon foil.

The foils used in this work were prepared (at Arizona Carbon Foil, Co.) by striking an arc between the sharpened tips of spectrographically pure graphite rods and collecting the ejected carbon atoms on a microscope slide. Electron diffraction measurements by Sander and Bukow [8] of as-deposited carbon foils prepared by the arc-evaporation method show two very diffuse rings, which is indicative of a very disordered or amorphous structure. However, this initial structure is modified by the bombardment of the foil by an ion beam, or by heating the foil either in an oven or with a laser. The bombardment of a carbon foil with a beam of energetic ions produces macroscopic changes in the structure and appearance of the foil that are readily apparent even to the naked eye. In addition, the ion-beam bombardment also causes microscopic changes in the structure and bonding of atoms in the bulk of the foil. These microscopic changes have been described by Sander, Bukow, and Buttlar [9] as a crystallographic transformation, or graphitization, of the foil.

In order to make an independent confirmation that ion-beam-induced graphitization was occurring in the foils used in this work, comparisons were made of the Raman spectra from as-deposited and proton-irradiated  $a$ -C foils. Typical Raman spectra for  $a$ -C foils used in this work are shown in Fig. 2. The as-prepared (unirradiated)  $2.0\text{-}\mu\text{g}/\text{cm}^2$   $a$ -C foil [Fig. 2(a)] exhibits a broad asymmetric peak from 1000 to

1800  $\text{cm}^{-1}$  with the highest peak at 1540  $\text{cm}^{-1}$  and a shoulder near 1350  $\text{cm}^{-1}$ . This spectral feature is a distinctive signature for amorphous carbon [10,11]. However, the Raman spectrum of the same foil following irradiation with a  $0.5\text{-}\mu\text{A}/\text{mm}^2$  beam of 25-keV protons to a total fluence of  $\sim 50$  mC [Fig. 2(b)] shows some obvious changes. The highest peak becomes narrower and shifts to around 1600  $\text{cm}^{-1}$ , while the relative intensity of the shoulder at 1350  $\text{cm}^{-1}$  increases. Comparing to the early work of Wada, Gaczi, and Solin [11], the Raman spectrum in Fig. 2(b) is very similar to that of their evaporated carbon film which had been annealed at 320°C for 59 h. Rouzaud, Oberlin, and Benybass [12] performed a detailed study of the graphitization process in evaporated carbon films. They heated samples under an inert gas flow from 25° to 2700°C and finally obtained polycrystalline graphite. They showed that the narrowing of the first-order Raman features and the enhancement of the peak at 1350  $\text{cm}^{-1}$ , as exhibited by the foils used in this work, is characteristic of the early stages of the graphitization process.

Hence the Raman spectroscopic study of the foils used in this work revealed that the ion-beam treatment induces an ordering of the originally disordered or amorphous structure. The overall effect is similar to that of annealing the film with the final result of a foil which has a higher degree of graphitic character.

#### IV. BALMER- $\alpha$ POLARIZATION MEASUREMENTS

In this section measurements of the tilt-angle dependence of the polarization of Balmer- $\alpha$  radiation emitted by beam-foil-excited hydrogen atoms are presented. This work is unique in that it represents measurements of the tilt-angle dependence of the Balmer- $\alpha$  Stokes parameters for the incident energy regime of 20–50 keV. The measurements presented in this section complement the work, performed at higher energies, of Liu, Gay, and Schüller [13] and Winter and Ortjohann [14] on the tilt-angle dependence of the Lyman- $\alpha$  radiation, and that of Singer, Dehaes, and Carmeliet [15], Berry *et al.*, [16] and Dehaes, Carmeliet, and Berry [17] on the linear polarization of the Balmer-series lines for normal incidence only.

A typical measurement of the tilt-angle dependence of the Stokes parameters for Balmer- $\alpha$  radiation emitted by beam-foil-excited hydrogen atoms is shown in Fig. 3. These data were taken with a 30-keV proton beam incident on a previously unirradiated spot of a  $1.5\text{-}\mu\text{g}/\text{cm}^2$  *a*-C foil. Some features of the data, which are common to nearly all beam-foil tilt-angle dependence measurements, can easily be explained by simple symmetry arguments. First, the linear polarization parallel and perpendicular to the beam,  $M/I$ , should not be dependent on the sign of the tilt angle. That is,  $M/I$  should be an even function of the tilt angle as was observed [see Fig. 3(a)]. Secondly, the linear Stokes parameter  $C/I$ , which is defined relative to axes which make angles of  $+45^\circ$  and  $-45^\circ$  with respect to the beam, should have odd symmetry [see Fig. 3(b)]. Finally, the circular Stokes parameter  $S/I$  should also have odd symmetry [Fig. 3(c)]. These symmetry arguments require the Stokes parameters  $C/I$  and  $S/I$  to be zero at normal incidence.

In addition, one can predict the sign of  $S/I$ , or the hand-

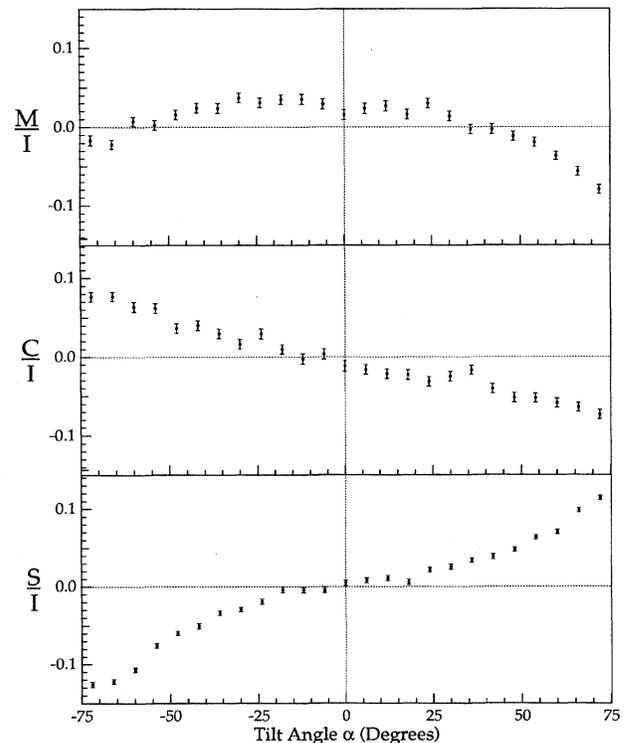


FIG. 3. A typical measurement of the tilt-angle dependence of the Stokes parameters for beam-foil Balmer- $\alpha$  radiation is shown. The data are for 30-keV protons incident on a  $1.5\text{-}\mu\text{g}/\text{cm}^2$  *a*-C foil.

edness of the circular polarization, from the following very simple model. Consider the beam-tilted-foil interaction from the perspective of the proton. As a proton emerges and recedes from the foil it “sees,” in its own rest frame, electrons from the foil surface moving past it in a direction opposite to its own motion. Due to the inclination of the foil, there will be an asymmetry in the electron density above and below the proton. For the case of a negative tilt angle, as is depicted in Fig. 1, there is a higher electron density surrounding the lower hemisphere of the proton as it recedes from the foil surface than its upper hemisphere. As a result, electrons are more likely to be captured into states where they will orbit the proton in a counterclockwise fashion rather than into states with clockwise orbits. More strictly, the probability for capturing electrons with a positive  $z$  component of angular momentum is higher than for capturing electrons with a negative value of  $L_z$ . Such states will subsequently emit left-hand circular polarization and give a negative value of  $S/I$ . In a similar manner one expects a positive value of  $S/I$  for a positive tilt angle. This is the customary tilt-angle behavior for the handedness of the circular polarization of radiation emitted by beam-tilted-foil-excited states and is in agreement with most experiments. Other models—for example, the density-gradient mode [18] which predicts a  $\sin(\alpha)$  dependence for the circular polarization—are in agreement with this anisotropic electron-capture picture.

Although the data of Fig. 3 are typical, not all the tilt-angle spectra presented in this paper can be described by the above simple models. In fact we observed that the polariza-

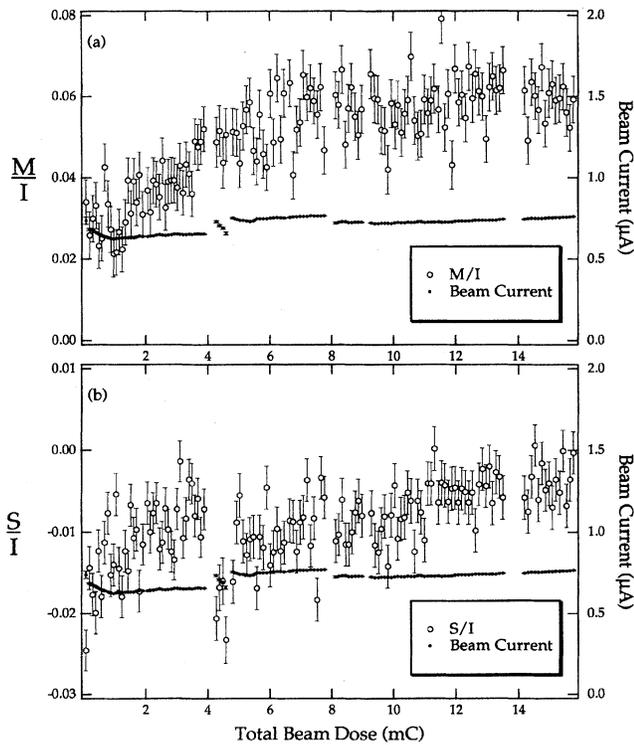


FIG. 4. The beam-dose dependence of the Stokes parameters (a)  $M/I$  and (b)  $S/I$  for a nearly constant beam-current density is shown. The data are for a 30-keV proton beam incident on a  $1.2\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil tilted at an angle  $\alpha = -21^\circ$ .

tion of Balmer- $\alpha$  radiation emitted by the foil-excited hydrogen atoms depended strongly upon the previous proton-irradiation history of the foil. This beam-fluence dependence, or dependence on the total accumulated beam dose, must be clarified before further tilt-angle dependence measurements can be presented. The linear Stokes parameter  $M/I$  and the circular Stokes parameter  $S/I$  were most strongly affected. Figure 4 shows (a)  $M/I$  and (b)  $S/I$  as a function of time for a nearly constant beam current of approximately  $0.65\ \mu\text{A}$ . Also shown in each figure is the beam current on the sample during the measurements. These data, taken with a 30-keV proton beam incident at  $\alpha = -21^\circ$  on a  $1.2\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil, were begun immediately after moving the beam to a previously unirradiated spot on the foil. In the subsequent discussions the degree of exposure to the proton beam is quantified by a measurement of the total accumulated charge collected on the foil sample. Since it was necessary to make these charge measurements without biasing the sample, the measured beam dose is influenced by the secondary-electron current. However, the charge measurements were all performed identically and thus serve as a valid point of comparison. The linear polarization represented by  $M/I$  starts out at a very small positive value, tends to increase with continued proton bombardment, and saturates at around 0.06 after an exposure of approximately 8 mC. The circular polarization starts out with a negative value as is expected for the negative tilt angle. However, as the foil is bombarded the degree of circular polarization increases toward zero, and it eventually

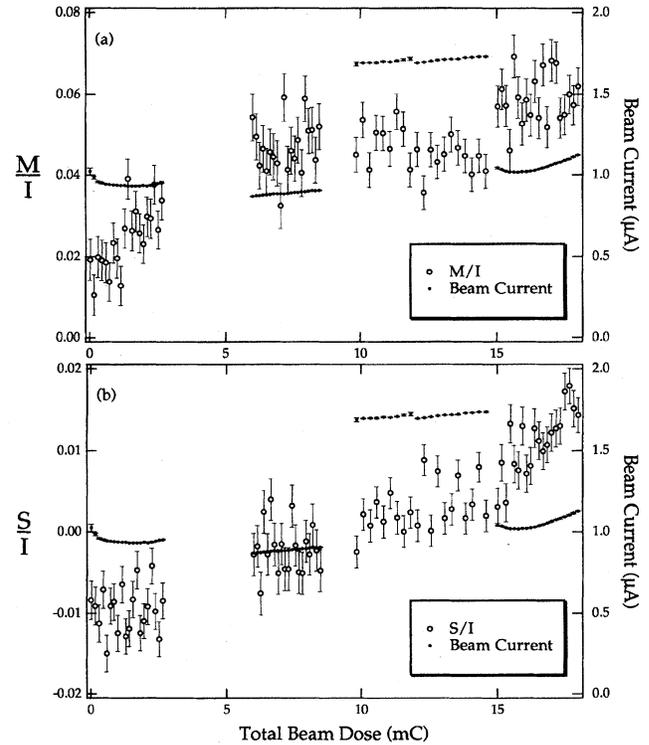


FIG. 5. The beam-dose dependence of the Stokes parameters (a)  $M/I$  and (b)  $S/I$  for different beam-current densities is shown. The data are for a 30-keV proton beam incident on a  $1.2\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil tilted at an angle  $\alpha = -21^\circ$ .

changes sign to give the *opposite* handedness of circular polarization from what is expected.

Gay and Berry [3] observed an increase in the alignment of  $2s\ ^1S-3p\ ^1P$  (5016 Å) He I radiation as a function of beam-current density which they attributed to a decrease in the secondary-electron yield at higher foil temperatures (i.e., at larger current densities). However, for our measurements of constant beam-current density such a temperature increase is unlikely. Rather the increase in the alignment is correlated with the beam-induced structure changes of the foil which were discussed in Sec. III. Figure 5 again shows time-dependent measurements of  $M/I$  and  $S/I$  for the Balmer- $\alpha$  radiation emitted following the transmission of 30-keV protons through a  $1.2\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil at an angle of incidence of  $\alpha = -21^\circ$ . In contrast to Fig. 4, the beam-current density was increased part way through the set of measurements. The increase in the beam-current density, which decreases the secondary-electron yield, did not drastically increase the alignment [Fig. 5(a)] as would be expected from the model of Gay *et al.* Also, the circular polarization continued to increase from its expected negative value and surprisingly passed through zero and became positive. After subsequently decreasing the beam current, the circular polarization did not decrease to its prior value, rather it continued its trend of becoming more positive. These facts suggest that the increases in  $M/I$  and  $S/I$  are due not to the instantaneous beam-current density but rather to the permanent changes in

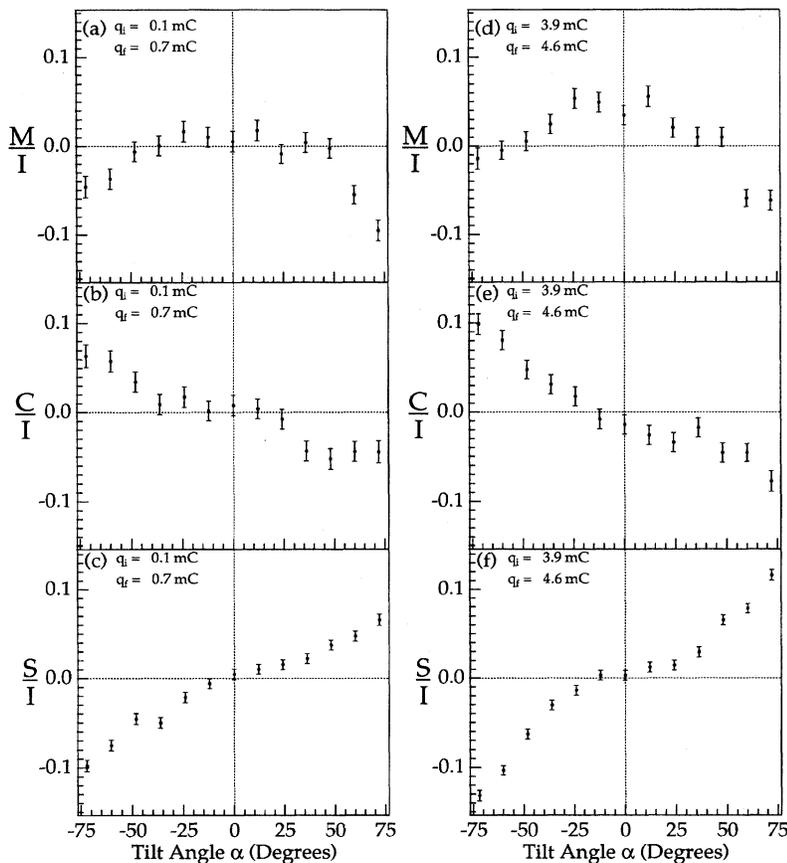


FIG. 6. Two measurements of the tilt-angle dependence of the polarization of Balmer- $\alpha$  radiation emitted by beam-tilted-foil-excited hydrogen atoms are shown. The spectra in (a)–(c) are for a 30-keV proton beam on a previously unirradiated spot of a  $1.5\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil. The total accumulated beam dose at the end of this run was 0.7 mC. The spectra in (d)–(f) are several runs later with the same beam at the same sample location. The total accumulated beam dose was 3.9 and 4.6 mC at the beginning and end of this run, respectively.

the structure of the foil induced by the continued proton bombardment.

The observed beam-fluence dependence significantly complicates the comparison of polarization measurements for different tilt angles or for different beam energies. In an effort to make such comparisons, we present tilt-angle dependence measurements at both extremely low beam fluence and at high beam fluence. To measure the tilt-angle spectra of the Stokes parameters at sufficiently low beam dose so that repeated spectra are not significantly different requires taking measurements more quickly and at fewer values of  $\alpha$  than was done for the data in Fig. 3. The measurements shown in Fig. 6 were taken as suggested above using a 30-keV,  $1\text{-}\mu\text{A}$  proton beam incident on a  $1.5\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil. The data in Figs. 6(a)–6(c) were taken immediately after moving the beam to a previously unexposed spot on the foil. The total accumulated beam dose at the end of this measurement was only 0.7 mC. The data in Figs. 6(d)–6(f) represent the seventh run in a series of runs on the same spot on the foil, each identical to the first. The beam dose at the beginning and end of this measurement was 3.9 and 4.6 mC, respectively. It must be emphasized that the beam current was kept nearly constant at  $1\text{ }\mu\text{A}$  during the entire time the foil was exposed to the beam, including the intermediate polarization measurements which are not shown in Fig. 6. Notice that the alignment at normal incidence is initially zero and only increases after increased proton bombardment. The behavior of the circular polarization is as expected with a negative value of  $S/I$  for negative tilt angles and positive  $S/I$  for positive tilt

angles. However, the dependence of  $S/I$  on tilt angle in Fig. 6(f) has changed from the initial linear relationship shown in Fig. 6(c). The degree of circular polarization at small tilt angles ( $|\alpha| \leq 30^\circ$ ) is decreasing (i.e.,  $|S/I|$  is approaching zero).

The trend in the evolution of the tilt-angle dependence of  $S/I$  hinted at by the data of Fig. 6 continues with increased beam dose. As with all the measurements presented in this paper, large irradiated beam doses were obtained by irradiating the foil at a constant beam-current density for a long period of time. During this long exposure it was possible to make many polarization measurements. The six plots of  $S/I$  versus tilt angle shown in Fig. 7 are a subset of 11 identical sequential measurements taken with a 30-keV,  $\sim 2\text{-}\mu\text{A}$  proton beam incident on a  $1.5\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil. The total accumulated beam dose at the beginning and end of each measurement is shown on the respective plot. The initial nearly linear dependence for  $S/I$  changes in a dramatic and unexpected way. For low tilt angles the observed sign of  $S/I$  is opposite to what one would predict from the anisotropic electron-capture model described earlier. The evolution of the  $\alpha$  dependence for the linear Stokes parameters for three of the 11 measurements is shown in Fig. 8. Notice the increase in the magnitude of  $M/I$  and the similarity in the shape of the  $C/I$  and  $S/I$  curves. Collectively these results indicate that either the state of the atom at the instant of electron capture, or the evolution of the excited state from the time of capture until the time of emission, or both, are different for the two cases of unirradiated and heavily

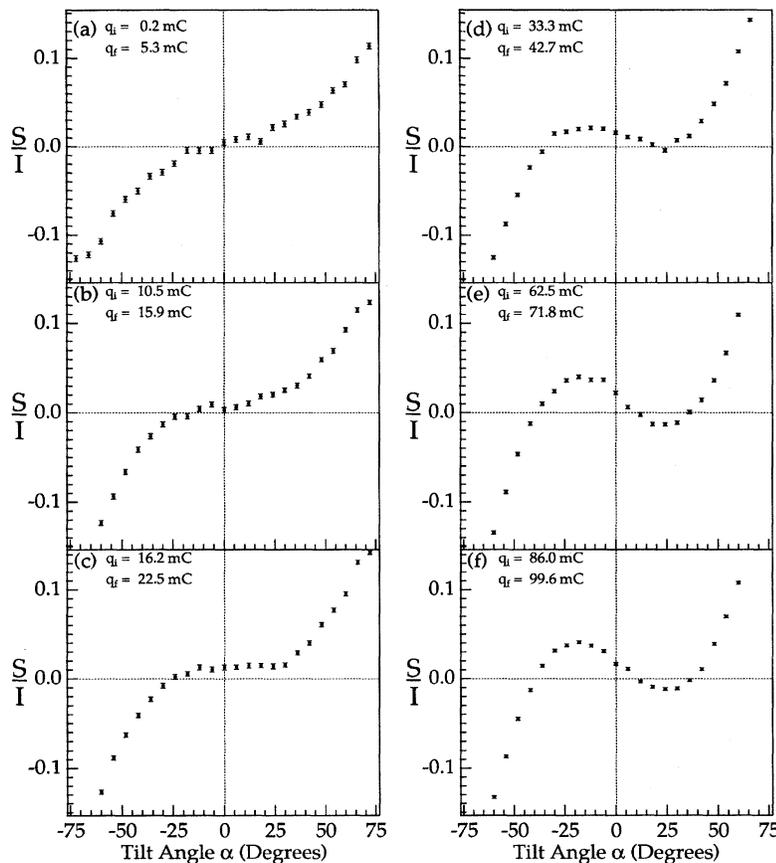


FIG. 7. A subset of several sequential measurements of the tilt-angle dependence of  $H\alpha$  circular polarization for 30-keV protons incident on a  $1.5\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil is shown. The tilt-angle dependence changes slowly from nearly linear in (a) to the anomalous, unexpected type of behavior in (f). The beam dose at the beginning and end of each run is shown on the respective plot.

proton-irradiated  $a\text{-C}$  foils. Presently, it is not possible by experiment to distinguish between these two atomic level descriptions. However, as we will show, the increase in the alignment and the unexpected, or anomalous,  $\alpha$  dependence of  $S/I$  are strongly correlated to the amount of irradiation by the proton beam which is known to change the properties of the carbon foil.

A series of tilt-angle spectra at beam energies of 25, 35, 40, and 45 keV were acquired using separate, distinct locations on a  $2.0\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil. The data shown in Fig. 9 are from the third in the series of runs at the different beam energies but nearly the same beam current. These measurements each represent nearly the same irradiation history, with the beam dose at the beginning of the run at approximately 10 mC and at the end approximately 15 mC. Comparison of the data in Fig. 9 shows that the anomalous behavior in the tilt-angle dependence of the circular polarization begins to develop earlier for lower energy projectiles. Also, at lower beam energies the amount of energy imparted to the foil by electronic energy loss events is greater, resulting in an enhanced graphitization of the foil. This observation supports the assertion that the anomalous  $S/I$  behavior is caused by the beam-induced graphitization of the foil.

Additional measurements were aimed at establishing that the graphitization of the foil was responsible for the observed beam-dose-dependent changes in the tilt-angle behavior of the Balmer- $\alpha$  polarization. This goal was accomplished by making measurements on foils which had been preannealed

by two different methods each independent of proton irradiation. The first method employed to graphitize the foil was  $\alpha$ -particle bombardment. A previously unirradiated spot of a  $1.4\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil was subjected to a 30-keV,  $\sim 1\text{-}\mu\text{A}$   $\text{He}^+$  beam for nearly 3.5 h. The total accumulated beam dose during the  $\alpha$ -particle irradiation was 9.6 mC. After the  $\text{He}^+$  treatment, the irradiated portion of the foil had become more graphitic. Next a measurement of the tilt-angle dependence of the Balmer- $\alpha$  Stokes parameters was made using a 30-keV proton beam incident on the  $\text{He}^+$ -irradiated spot. The resulting tilt-angle spectra were similar to those from the proton-irradiated foil surfaces shown earlier [see Figs. 8(a), 8(d), and 7(c)]. The second method used to graphitize the originally amorphous carbon foils was heating of the foil in vacuum. Measurements were made of the tilt-angle spectra of the Stokes parameters of Balmer- $\alpha$  radiation emitted following the transmission of 30-keV protons through a heat-treated  $1.4\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil. The measurements were made after annealing the foil to a temperature of  $325^\circ\text{C}$  for 2 h, and then again after a subsequent annealing to  $500^\circ$  for 2 h. The data from the hotter heat treatment begin to show the characteristics of the heavily proton-bombarded foils. Unfortunately, heat treatments at temperatures in excess of  $500^\circ\text{C}$  were not possible.

## V. CONCLUSIONS

In this paper we present measurements of the tilt-angle dependence of the Stokes parameters for Balmer- $\alpha$  radiation

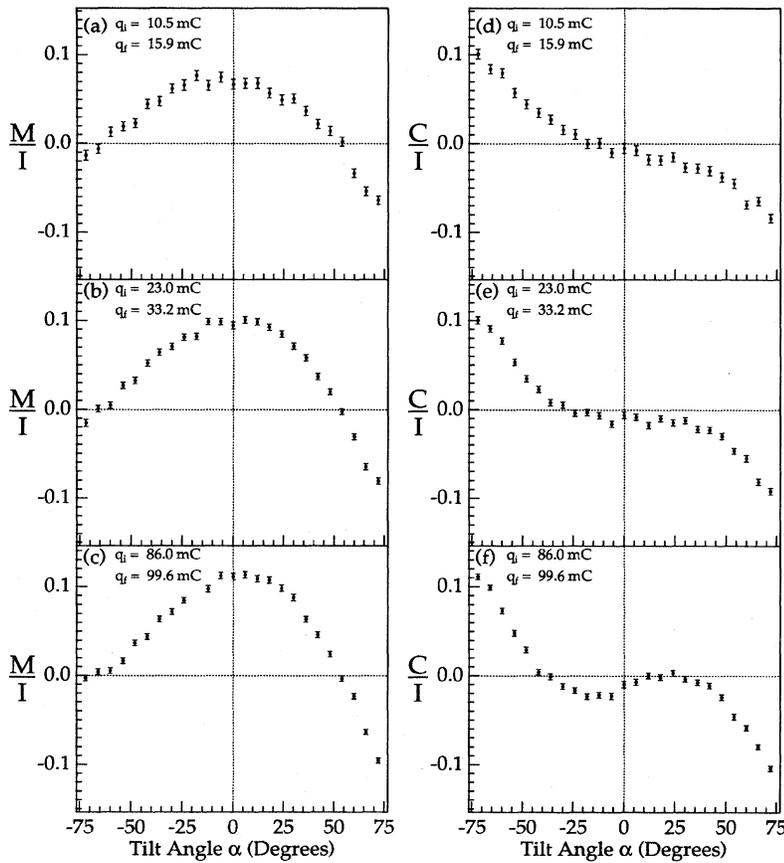


FIG. 8. A subset of several sequential measurements of the tilt-angle dependence of linear Stokes parameters  $M/I$  (a)–(c) and  $C/I$  (d)–(f) of Balmer- $\alpha$  radiation emitted following the transmission of 30-keV protons through a  $1.5\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil is shown.

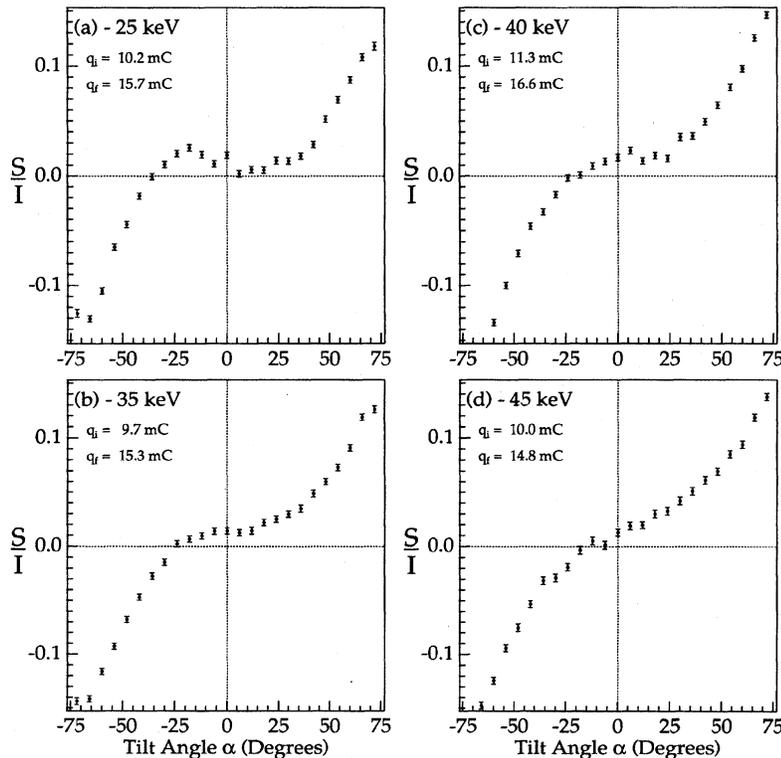


FIG. 9. The tilt-angle spectra of the linear Stokes parameter  $S/I$  for similar irradiation histories are shown for (a) 25-keV, (b) 35-keV, (c) 40-keV, and (d) 45-keV protons incident on a  $2.0\text{-}\mu\text{g}/\text{cm}^2$   $a\text{-C}$  foil. These measurements each represent nearly the same irradiation history, with the beam dose at the beginning of the run at approximately 10 mC and at the end approximately 15 mC. The anomalous  $S/I$  behavior has developed sooner at the lower beam energies.

emitted by beam-foil-excited states following the transmission of (20–50)-keV protons. The polarization data show a strong dependence on the prior history of exposure of the foil to the proton beam. This beam-fluence dependence changes the shape of the tilt-angle dependence of the Stokes parameters. Specifically the degree of alignment, indicated by the linear Stokes parameter  $M/I$ , is enhanced and the circular Stokes parameter  $S/I$  changes from an initially well-understood linear-type behavior to a behavior which, for low tilt angles, gives the opposite handedness of circular polarization from what is expected. These changes in the tilt-angle dependence of the Stokes parameters have been systematically correlated with the beam-induced graphitization of the foil which is known to occur from Raman measurements. The following observations support this assertion: the enhancement of  $M/I$  and the anomalous behavior of  $S/I$  occur (1) after prolonged exposure to a constant current-density proton beam, (2) more rapidly for lower energy proton beams which cause a more rapid rate of graphitization, (3)

after preirradiating (graphitizing) a spot on the carbon foil with a  $\text{He}^+$  beam, and (4) after graphitizing the foil by heating it in vacuum. *The evidence suggests that graphitization may be brought about both by increasing the sample temperature and/or by low-intensity, dose-dependent, proton bombardment, more or less independent of the local temperature.*

The changes in the tilt-angle dependence of the Stokes parameters, although correlated with the graphitization of the foil, remain to be connected to the details of the electron-capture process. Specifically, it is unknown whether the differences in the observed Stokes parameters for unirradiated and graphitized carbon foils are due to electron capture into a different excited-state configuration or due to electric-field-dependent quantum-mechanical phase evolution of the excited states between the times of creation and deexcitation. Further investigative efforts are required to develop a better understanding of this issue.

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