

Material-dependent Variations of Alignment in Beam-Foil Excitation

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Abstract

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We have measured the linear polarization fractions of the $2s\ ^1S-3p\ ^1P$ He I transition at $5\ 016\ \text{\AA}$ excited by the passage of helium ions through thin perpendicular foils at beam energies of 40 to 220 keV. Metallic foils such as Au and Ag show less polarization than carbon, while non-conducting SiO_2 foils produce increased linear polarization. Time variations of the polarization show that the build-up of impurities on the final surface of the foils yields carbon-foil like polarizations. At higher beam energies of 0.6 to 1.0 MeV, all foils show increased polarization for neon transitions, but decreased polarization for He II transitions with respect to the carbon foil values.

1. Introduction

The production of atomic alignment in beam-foil excitation leads to several techniques in atomic structure measurements; for example, zero field quantum beats to measure fine and hyperfine structures, level crossing resonances and g -value beats [1]. The degree of alignment generally limits the accuracy of such experiments. Also, it has been found experimentally that at useful beam energies, only the light ions have significant alignments leading to polarization of above 10%. Some heavier ions, notably the rare gases, have shown linear polarization fractions of 5% to 10%².

All the above types of experiments have used carbon foil excitation. In this paper, we investigate the dependence of the production of alignment on the exciter foil material, with two objectives in mind: materials which give larger alignments would be useful in atomic structure measurements and the variation of alignment with foil material should help to analyze the interaction mechanism leading to the alignment production.

Berry et al. [3] found that both alignment and orientation vary with the tilt angle of the final surface of the exciter foil with respect to the ion beam axis. These results show that the alignment and orientation depend on the final surface interaction of the ions as they leave the foil. Consequently, in experiments involving different foil materials, it is important to know the cleanliness of the final surface. Therefore, we have also studied the time variations of alignment during build-up of hydro-carbon impurities on the foil surface.

2. Experiment

We accelerated $^4\text{He}^+$ ions at energies between 40 keV and 220 keV and after magnetic selection they passed through the standard

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beam-foil excitation system shown in Fig. 1. Up to 20 foils on a foil wheel could be included in the target chamber, and rotated individually into the beam. The light emitted from the foil-excited ions was detected at close to 90° to the beam axis. The polarization state of the emitted light was measured through a rotatable phase plate (near to a quarter-wave) and a fixed linear polarizer. A McPherson 1/3 m—218 monochromator, a Bailey Centronic 4249 photomultiplier and photon counting electronics completed the detection system. Normalization of photon yield to beam charge collected at the Faraday cup was used. The polarization analysis system has been described elsewhere [4].

The target chamber vacuum was maintained by a liquid nitrogen trapped oil diffusion pump at a value of 4 to 8×10^{-7} torr.

The observations to be described were mostly made on the $5\ 016\ \text{\AA}$, $2s\ ^1S-3p\ ^1P$ transition of He I. The transition was well resolved by the monochromator from the nearest He I transition at $5\ 047\ \text{\AA}$, and the second order $2\ 511\ \text{\AA}$ He II transition was not transmitted by the polarization system.

3. The polarization system

Since the foil is always perpendicular to the beam axis, the excitation system has cylindrical symmetry, and the excited states may be aligned but not oriented. Moreover, only one alignment parameter, which compares the angular momentum with respect to the beam-axis direction to any other direction in the foil plane, is non-zero [5, 6]. This single non-zero alignment parameter, the A_0^{col} Fano-Macek parameter [7], or σ_0^2 the density matrix component taking the beam axis as the z -axis [6], is measured by the linear polarization fraction

$$\frac{M}{I} = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}} \quad (1)$$

where I_{\parallel} and I_{\perp} are the intensities of light emitted in the observation direction (Fig. 1) linearly polarized parallel and perpendicular to the beam axis. For the transition with angular momentum quantum numbers $F \rightarrow G$ we have [6, 7]

$$\frac{M}{I} = \frac{-3H_2(FG) \cdot \sigma_0^2}{2\sqrt{2}\sigma_0^0 - H_2(FG)\sigma_0^2} = \frac{3h^{(2)}(FG)A_0^{\text{col}}}{4 + h^{(2)}(FG)A_0^{\text{col}}} \quad (2)$$

where H_2 and $h^{(2)}$ are ratios of $6-j$ coefficients defined in refs. [6] and [7].

In the polarization system of Fig. 1, the transmission axis of the polarizer was set perpendicular to the beam direction. Then, if the phase plate has a retardation angle of δ , the light transmitted through the system as a function of the angle β of the fast axis of the phase plate to the beam direction can be written

$$I(\beta) = 1 + \eta \cos 4\beta \quad (3)$$

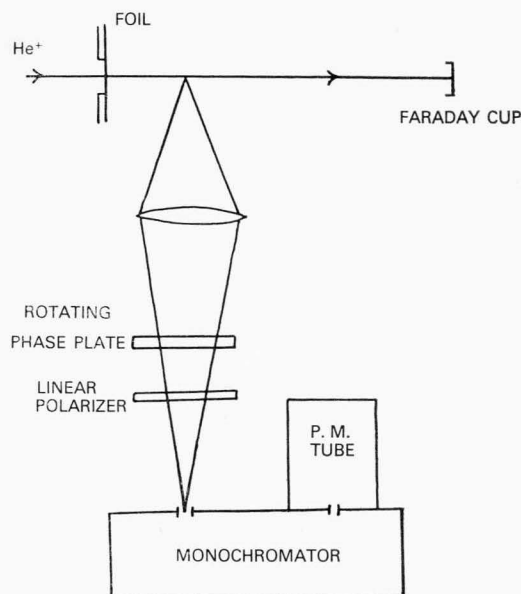


Fig. 1. The experimental arrangement. The viewing region was a few mm from the foil surface.

The polarization fraction can be calculated from the modulation ratio η using

$$\frac{M}{I} = \frac{2\eta}{(1 - \cos \delta) + (1 + \cos \delta)\eta} \quad (4)$$

Measurements were made at phase plate angles 0° , 45° , 90° etc.

4. Results

4. Time variations of polarization fractions for C, Au and Ag foils

A major problem in studying the characteristics of clean metallic surfaces in vacuum is the deposition rate of impurities from the residual gases. Oxygen and water probably contaminate the Au and Ag surfaces when placed in the target chamber.

A greater problem arises from the coverage of hydrocarbons which are present in our background vacuum of 5×10^{-7} torr. Even with a liquid nitrogen trapped diffusion pump these impurities produce a monolayer coverage within a short time. Subsequently, ion beam bombardment "cracks" the hydrocarbons, leading to carbon build-up on both the front and back surfaces of the foils. This carbon build-up was observed in electron microscope photographs on all foils after a few minutes of helium ion beam bombardment (by courtesy of Prof. M. Isaacson, University of Chicago).

We measured the time dependence of the linear polarization fraction, M/I of the $5\ 016\ \text{\AA}$ He I transition $2s\ ^1S-3p\ ^1P$ for carbon, gold and silver foils newly placed in the vacuum. The time resolution was approximately 50 seconds, the limitation being to obtain sufficient photon counts to reduce statistical errors in M/I to about ± 0.02 . Measurements were taken over times of up to 10^4 s at beam energies between 80 and 200 keV. The results for the carbon foils were constant in time. The linear polarizations observed using the Au and Ag foils varied in time and we show two examples in Fig. 2. The linear polarization tends to the carbon value with a time constant of about 1 500 s. All the time variation curves could be fit to the form

$$\frac{M}{I}(t) = M_0 + (M_c - M_0)(1 - e^{-t/\tau}) \quad (5)$$

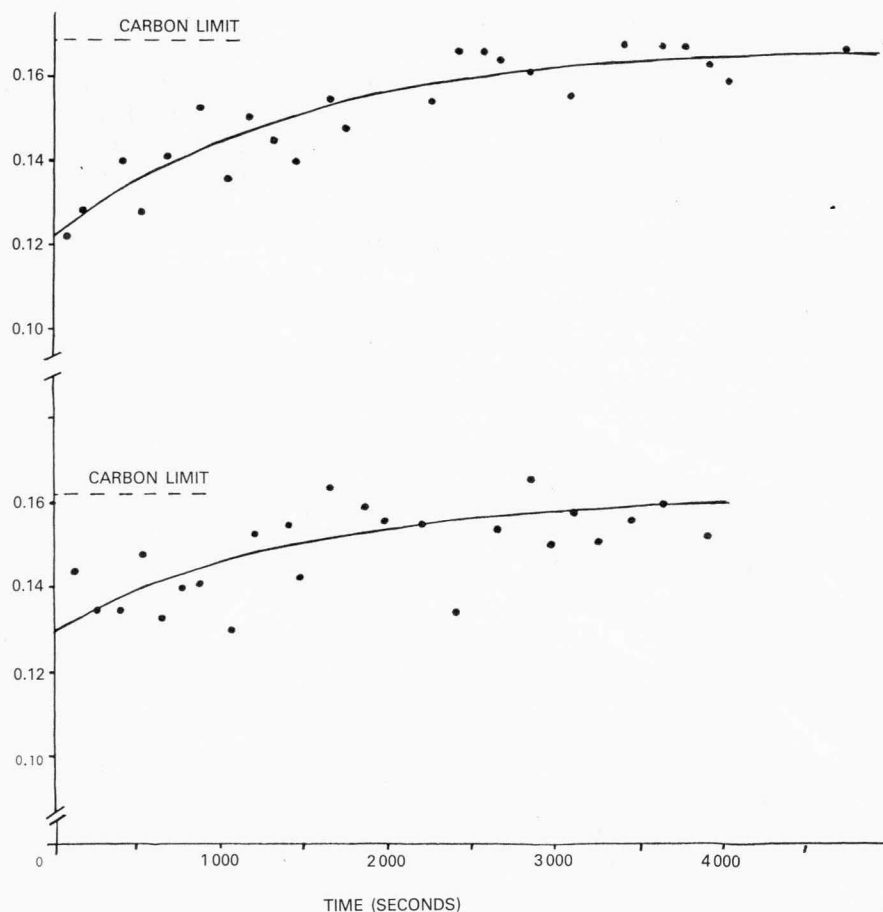


Fig. 2. Variation in time of the linear polarization fraction M/I of He I $5\ 016\ \text{\AA}$ excited by a thin (a) gold, and (b) silver foil as a function of bombardment time. The incident ion beam energy is 129 keV and a beam current of $2\ \mu\text{A}$ over $1/4''$ diameter. The solid line is a fit to eq. (5) and the parameters are for (a) $M_0 = 12.2$, $M_c = 16.8$, $\tau = 1\ 500$ s, and (b) $M_0 = 13.0$, $M_c = 16.2$, $\tau = 1\ 500$ s.

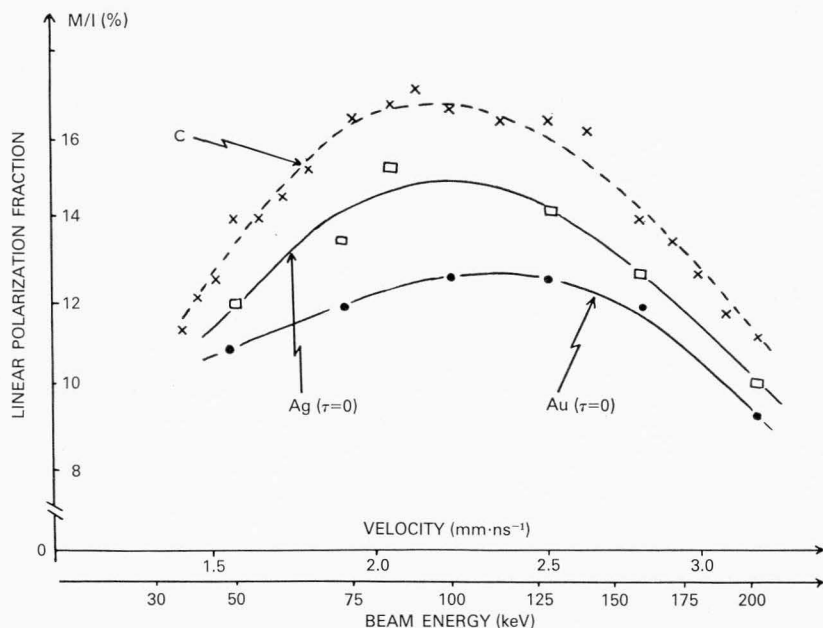


Fig. 3. Variation of the linear polarization fraction M/I of He I 5 016 Å with ion beam energy for carbon, gold and silver foil excitation.

where M_0 is the initial polarization from the Au or Ag surface, M_c is the equilibrium carbon value, and τ is the time constant.

The time constant τ was smaller for higher background pressures in agreement with its being a function of the deposition rate of cracked hydrocarbons on the foil surface. The time required to obtain the carbon value of the linear polarization fraction M/I is surprisingly long. In 10^3 s the surface may be coated with many layers (10 or more) of impurities, but the measured polarization fraction suggests that the excitation of the outgoing ion is still dependent on the metallic (Au or Ag) part of the foil. Thus, either there is a long range (many atomic layers) memory in the final surface interaction, or the carbon build-up rate is much slower, with a single monolayer coverage taking about 10^3 s.

The Au and Ag foils were generally 2-component foils of $5 \mu\text{g cm}^{-2}$ of carbon and $10 \mu\text{g cm}^{-2}$ of the metal. For reversed foils with the beam passing through first the metal and then the carbon part, polarization fractions corresponding to excitation from carbon foils were always obtained.

4.2. Energy dependence

In Fig. 3 we compare the linear polarization fractions for the 5 016 Å $^4\text{He II } 2s^1S-3p^1P$ transition excited by carbon, gold and silver foils over the beam energy range of 40 to 200 keV. The maximum alignment occurs near 110 keV for all the foils and the carbon values are close to those observed previously by Berry and Subtil [8]. The small discrepancies between the results of ref. [8] and those of Fig. 3 are due to the difference in thickness of the carbon foils—those used in ref. [8] were $\geq 10 \mu\text{g cm}^{-2}$, compared with $5 \mu\text{g cm}^{-2}$ in this study [9]. Also, the carbon foil induced alignment is dependent on beam current density [9], which was not noted in ref. [8], but was maintained constant at $3.2 \mu\text{A cm}^{-2}$ in this experiment. The results shown for the Au and Ag foils are the values obtained with fresh foils, using extrapolation to $t=0$ of the beam bombardment.

4.3. Variation of the gold foil thickness

Carbon foils of $5 \mu\text{g cm}^{-2}$ thickness were coated with very thin layers of gold as measured during evaporation. In Fig. 4 we show that the linear polarization fraction M/I for the 5 015 Å transition approaches the gold equilibrium value when a thickness of about $5 \mu\text{g cm}^{-2}$ is reached and remains constant for larger

thicknesses. Church and Michel [10] have found that polarization fractions of the He I 4 922 Å transition $2p^1P-4d^1D$ vary for tilted foils with gold thicknesses between 5 and $10 \mu\text{g cm}^{-2}$. However, their results may be due to the time dependent effects we discuss above. They suggest that the variations may be due to non-uniform coverage of the carbon foil by the gold layers. This is probably occurring in the very thin gold layers used in this experiment, since $1 \mu\text{g cm}^{-2}$ corresponds to only 2 gold mono-layers.

4.4. Time variations of polarization fractions from SiO_2 foils

The low polarizations observed in He I 5 015 Å from the metallic Au and Ag foils (Fig. 3) suggest that non-conducting foils might enhance the polarization production. Consequently, we have measured the polarization of 5 015 Å using thin foils of $5 \mu\text{g cm}^{-2}$ carbon backed with $25 \mu\text{g cm}^{-2}$ of SiO_2 . The time dependence of M/I for three different beam current densities is shown in Fig. 5. The curves can all be fit to an equation of the form

$$\frac{M}{I}(t) = M_C + M_L e^{-t/\tau_L} - M_S e^{-t/\tau_S} \quad (6)$$

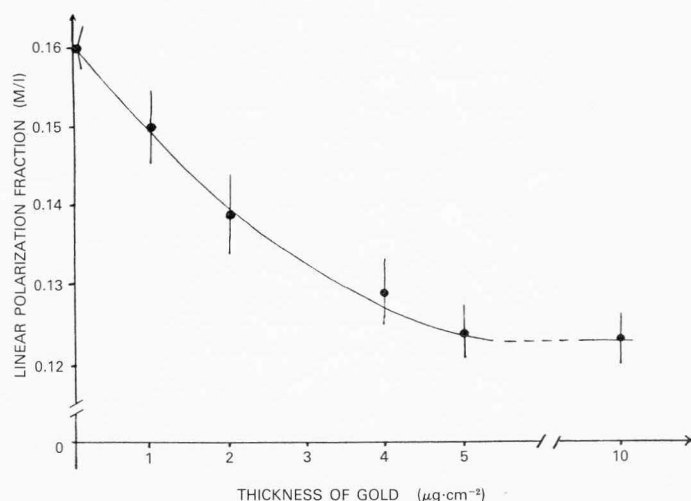


Fig. 4. Variation of the linear polarization fraction M/I of He I 5 016 Å with the thickness of a final gold layer supported on a $5 \mu\text{g cm}^{-2}$ carbon foil. The incident ion beam energy was 110 keV with a beam current of $5.6 \mu\text{A cm}^{-2}$.

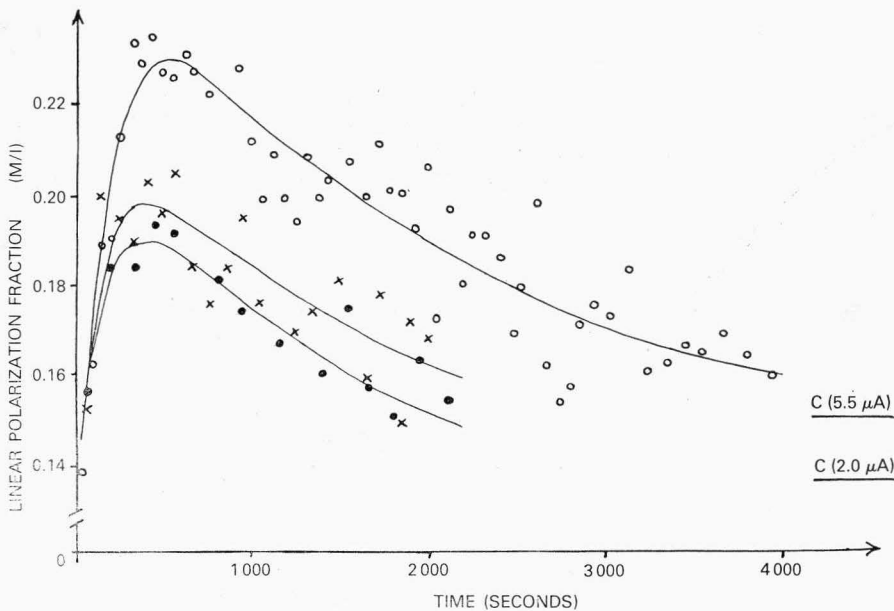


Fig. 5. Variation in time of the linear polarization fraction M/I of He I 5 016 Å excited by a $25 \mu\text{g cm}^{-2}$ SiO₂ foil (supported on $5 \mu\text{g cm}^{-2}$ carbon). The three sets of data are for beam current densities of $5.6 \mu\text{A cm}^{-2}$ (●), $11.2 \mu\text{A cm}^{-2}$ (×) and $25.2 \mu\text{A cm}^{-2}$ (○) at a beam energy of 161 keV. The solid line fits are eq. (6) using the parameters given in Table I.

where M_C is a linear polarization fraction for a thin carbon foil at the same beam current density, and $M_L + M_C$ is a linear polarization fraction which we suggest is associated with a clean SiO₂ surface. τ_L and τ_S are relaxation times: τ_L corresponds to the slow decrease in $M/I(t)$ to the carbon limit, and τ_S corresponds to the rapid increase at small times.

We have made best fits to the data of Fig. 5 in terms of the parameters M_C , M_L , M_S , τ_L and τ_S with the results shown in Table I. We suggest that the rapid initial rise in the fractional polarization M/I is due to the removal of surface impurities such as oxygen, nitrogen and water, while the steady accumulation of cracked hydrocarbons then leads to the reduction of M/I to the carbon-foil limit. A possible alternative explanation is an initial change in structure of the SiO₂ surface leading to the increased S/I , followed by the same cracked hydrocarbon build-up. However, no evidence for structural changes could be seen in electron microscope scans.

Eq. (6) can be derived assuming that the contaminant removal rate is proportional to the amount of contaminant remaining and the carbon coverage rate is proportional to the fractional hydrocarbon-free area. The carbon build-up rates observed by Dumont et al. [11] of $1 \text{ ng cm}^{-2} \text{ min}^{-1}$ for D⁺ beams in 10^{-6} torr vacuum correspond to a monolayer coverage time of 3 000 seconds. Hence, our observed polarization variations appear to be sensitive to single monolayer build-up in the foil surface. For the case of $M_L = M_S$, the initial surface contaminants produce an M/I equal to the carbon value M_C . This last assumption is consistent with the initial contaminated carbon, clean carbon, and cracked hydrocarbon surfaces all leading to the same M/I . This assumption is consistent with the Au and Ag results of Fig. 2 only if we also assume that such contaminants are negligible or removed within

Table I. The time dependent polarization of He I, 5 016 Å, $2s^1S-3p^1P$, at 161 keV beam energy excited by a thin SiO₂ foil fit to eq. (6), $M/I(t) = M_C + M_L e^{-t/\tau_L} - M_S e^{-t/\tau_S}$

The fits derived from these parameters are shown in Fig. 5

Beam current M_C ($\mu\text{A cm}^{-2}$)	M_L	M_C	τ_L (s)	τ_S (s)
25.2	0.142	0.122	2 100	181
11.2	0.135	0.09	1 680	162
5.6	0.130	0.09	1 400	168

a few seconds for Au and Ag surfaces. We conclude that the polarization produced by clean SiO₂ surfaces is significantly higher than that produced by C, Ag and Au.

4.5. High energy results

Similar time dependence measurements were made at higher incident beam energies of 0.6 to 2 MeV at the Argonne Dynamitron accelerator. The Ne II 3 568 Å $3s^1 2D_{5/2}-3p^1 2F_{7/2}$ transition has a linear polarization fraction M/I for carbon foil excitation of 0.045 at 0.6, 0.8 and 1.0 MeV. Au, Ag and SiO₂ foils all gave an initial M/I for this transition of between 0.06 and 0.07 decaying exponentially to the carbon value of 0.045 within a few thousand seconds at all three beam energies. The percentage variation of M/I for the Ne III 2 613 Å transition was similar (0.065 decaying to 0.05) with foils of Au, SiO₂ and WO₃. However, at 0.6 MeV energy, the He II 3 204 Å $n=3-5$ transition showed low initial M/I for foils of Au (0.06), SiO₂ (0.045), Li₂O₃ (0.045) and WO₃ (0.02) rising to a carbon value of 0.073 ± 0.005 .

In Fig. 6 we show representative results for the polarizations decreasing in time for the Ne II 3 568 Å and Ne III 2 613 Å transitions, and increasing in time for the He II 3 204 Å transition. The decay time constant for the approach to carbon foil equilibrium is approximately 2 000 seconds in all cases (for a background vacuum of 5×10^{-7} torr).

The WO₃ foils produced a more complex time dependence of the polarization of the He II transitions $n=3-4$ at 4 686 Å, and $n=3-5$ at 3 204 Å. We show in Fig. 7 the results for 3 204 Å at 0.6 MeV beam energy. A reasonable two exponential fit can be obtained with a slow decay time of about 2 000 s (2 140 s by a fitting procedure) and a fast decay of 200 s. It should be noted that the fast and slow decay times are in good agreement with those obtained with SiO₂ foils for the polarization of 5 016 Å of He I. This suggests that the mechanisms involved are similar: an initial fast removal of impurities such as N₂, O₂ and H₂O followed by the slow build-up of "cracked" hydrocarbons.

5. Discussion

At low beam energies, the reduced polarizations of 5 016 Å obtained using Au and Ag foils suggest that better-conducting foils lead to lower polarizations. This is verified by the observation that the non-conducting SiO₂ foil enhances the linear polarization

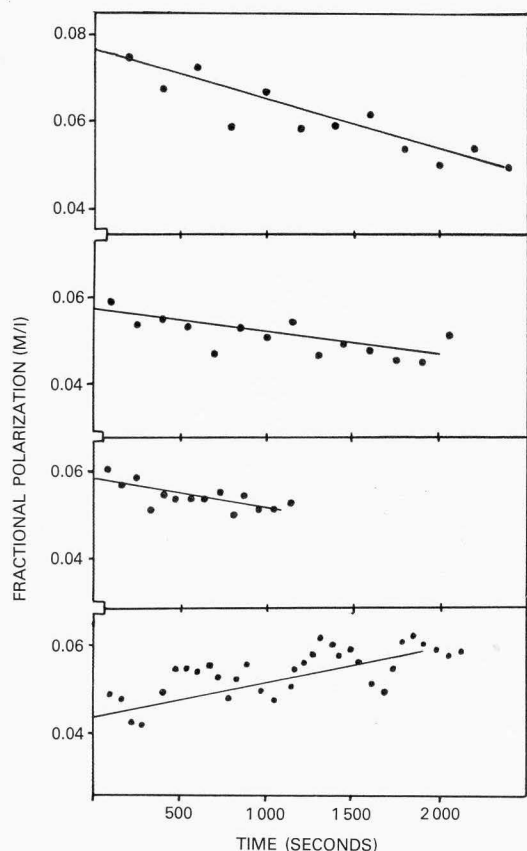


Fig. 6. Time dependence of the linear polarization fractions M/I for (a) Ne III, 2.613 Å, at 1 MeV with WO_3 foil, (b) Ne II, 3.568 Å, at 0.8 MeV with Au foil, (c) Ne II, 3.568 Å, at 0.8 MeV with SiO_2 foil and (d) He II, 3.204 Å, at 0.6 MeV with Li_2O_3 foil.

fraction. However, silver has a slightly higher electrical conductivity than gold (6.21 compared with $4.55 \times 10^9 \text{ ohm}^{-1} \text{ cm}^{-1}$), whereas the M/I obtained with Ag foils is intermediate between Au and C.

The excited state of the helium projectile is formed at the final surface of the foil, and electron capture may be the dominant mechanism. Trubnikov and Yavlinskii [12] have calculated the recombination probability for electron tunneling from a square

well with Fermi energy E_F and work function W to hydrogenic bound states. Schröder and Kupfer [13] have used this model to calculate M/I for the $2p$ state of hydrogen, and obtain a value of 1.0, the maximum possible. This result is much higher than observed for any beam-foil excited transitions. Other mechanisms such as Auger processes contribute to the final excited state populations, but this static model of the surface interaction is clearly inadequate. The strongly disruptive forces of the fast moving ion change the foil characteristics exactly where the interaction takes place. The material polarization in the particle wake¹⁴ is one example of such dynamic effects. Gold and silver have the same Fermi energy and work function and would show the same polarization in the static model. It is clear from Fig. 3 that they do not, and dynamic effects must be included.

Comparison of the results at high energy (0.6 to 1.0 MeV) to those at low energy (40 to 220 keV) shows that the material dependence of the polarization fraction changes with ion energy. Two points should be emphasized: (1) For a given foil (e.g. Au, Ag or SiO_2), at a given energy, the polarization fraction is higher for one ion (e.g. Neon transitions), but is lower for a different ion (e.g. He II), than the carbon values; and (2) SiO_2 foils give higher polarization for 5.016 Å He I at low energy (30–200 keV) but give lower polarization for 3.204 Å He II at higher energy (0.6 to 1.0 MeV), than the carbon values.

The two-component time variations of polarization for the SiO_2 and WO_3 foils seem to be understood. However, it is puzzling that two-component curves are not found for all foil materials. The initial impurities on the SiO_2 and WO_3 foils give strongly different polarization changes. Thus, the impurities either are removed in a few seconds in the metallic foils, or they do not produce a radical change in polarization for such an environment.

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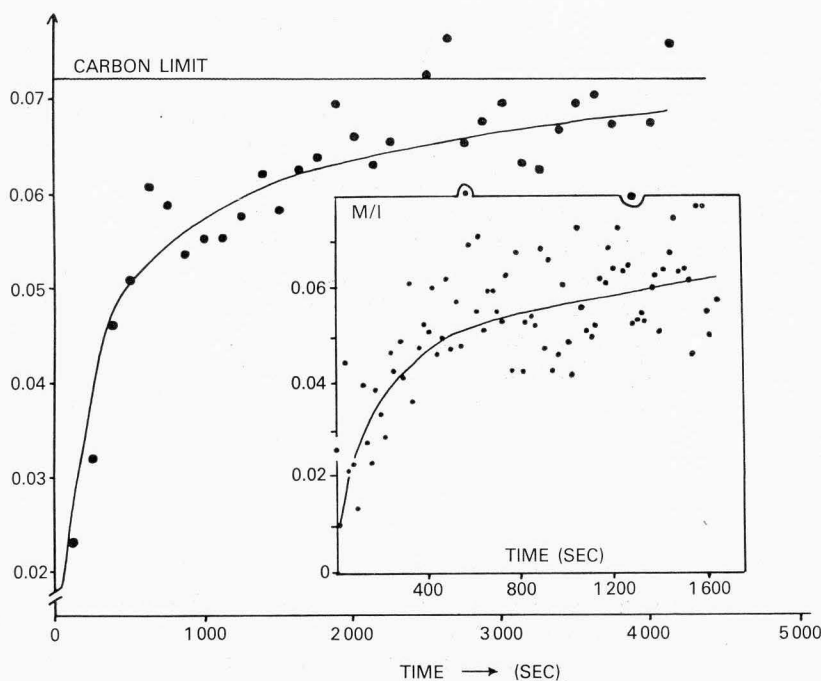


Fig. 7. Variation in time of the linear polarization fraction M/I of He II 3.204 Å, $n=3-5$, excited by a WO_3 foil. The solid line fit is the curve $M/I(t) = 0.073 - (0.025e^{-t/200} + 0.040e^{-t/2140})$. The inset shows more detailed results near $t=0$.

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