Reprinted from: BEAM-FOIL SPECTROSCOPY, VOL. 2 (1976)
Edited by Ivan A. Sellin and David J. Pegg
Book available from: Plenum Publishing Corporation
227 West 17th Street, New York, New York 10011

THE SURFACE INTERACTION IN BEAM FOIL SPECTROSCOPY

H. G. Berry

Department of Physics, University of Chicago, Chicago, Ill. 60637

and Argonne National Laboratory, Argonne, Ill. 60439

and

L. J. Curtis, D. G. Ellis, R. D. Hight, and R. M. Schectman

Department of Physics and Astronomy, University of Toledo, Toledo, Ohio 43606

Abstract

We review the measurements of the changes in light polarization in the beam-foil source when the foil tilt angle is varied. Comparisons are made with theories of the final surface interaction.

1. Introduction

The passage of fast heavy ions through solids has yet to be described in terms of an accurate theoretical model. Some progress has recently been made on the understanding of the effective charge states of the moving ions, both experimentally[1] and theoretically,[2] but the states of binding of the outer shell electrons remain essentially unknown. In particular, possible variations of such effective charge and excitation states with differing solids have not been measured or predicted. Following the discovery of atomic alignment in the beam-foil light source[3] through the observations of linearly polarized quantum beats in field-free radiative decays, it was natural to investigate whether alignment measurements could give information on the states of excitation of heavy ions in solids. We discuss below some of the progress made in this direction since the last beam-foil conference.

The first necessary step is to describe the state of atomic alignment produced in the beam-foil excitation mechanism in terms of the (observed) light emitted in the radiative decay of the excited state. Ellis,[4] and Fano and Macek[5] have thus related the excited state density matrix or state multipole moments to the polarization and angular distribution of the emitted radiation. We briefly discuss how symmetry conditions of the excitation can be used to predict the various possible polarizations in the emitted light.

For a spherically symmetric source, the radiation is emitted isotropically and is unpolarized, and the source can be described by a single parameter – the number of excited atoms N. In quantum mechanical terms, since no direction is specified, all different angular momentum sub-levels are equivalent and we have statistical populations.

In Fig. 1, we show successive reductions in the symmetry of the excited light source which lead to the need for more parameters to adequately describe the source, which in turn affect the emitted radiation. In Fig. 1(b) is a cylindrically symmetric source, such as electron beam excitation of a gas. The $\hat{z}$-axis now differs from the $\hat{x}$ and $\hat{y}$ directions, and a second parameter, <u>the alignment</u>, is introduced. The cross-sections to sub-levels of different $|m_L|$ may now be different, and Percival and Seaton[6] have related these cross-sections to the fractional linear polarization of the emitted light. For example, for a $^1P \rightarrow {}^1S$ transition, the z-axis as direction of quantization, then the fractional linear polarization of light observed perpendicular to the z-axis is

$$P_L = \frac{I^{\parallel} - I^{\perp}}{I^{\parallel} + I^{\perp}} = \frac{\sigma(m = \pm 1) - \sigma(m = 0)}{\sigma(m = \pm 1) + \sigma(m = 0)} \quad \ldots\ldots \; . \qquad (1)$$

Until two years ago, the beam-foil light source was considered to be such a source, describable in terms of two parameters (N, P_L) for each excited state. One additional important quality is that all excitations occur at a time t=0 on the z-axis defined to better than 10^{-14} sec, which gives rise to the zero field quantum beats, an example of which is shown in Fig. 2. Basically, we have produced a state which is not an energy eigenstate of the free atom Hamiltonian.

However, the beam-foil interaction at z=0 also depends on the <u>direction</u> of the beam velocity $\vec{v}$. Thus, Eck[7] pointed out that the interaction need not be invariant under reflection in the x-y plane of the foil. He proposed a simple test of comparing the Lyman α decays of n=2 hydrogen in an electric field parallel and anti-parallel to the +z-axis. A phase shift of the electric-field induced Lamb shift quantum beats between $2s_{\frac{1}{2}}$ and $2p_{\frac{1}{2}}$ indicating that a

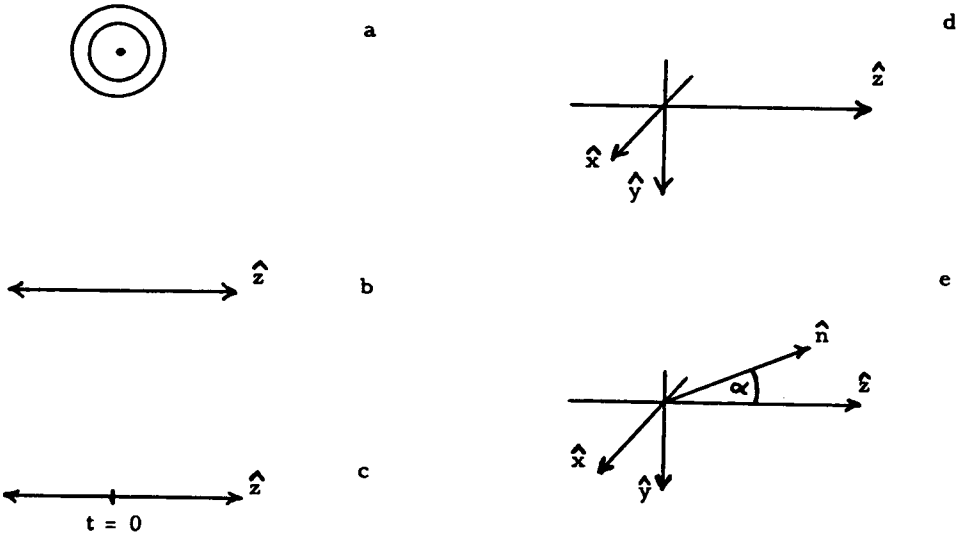

Fig. 1. Excitation source symmetries. a - spherical symmetry, b - cylindrical symmetry, c - excitation at t = 0, d - reflection assymmetry in x-y plane, e - loss of cylindrical symmetry.

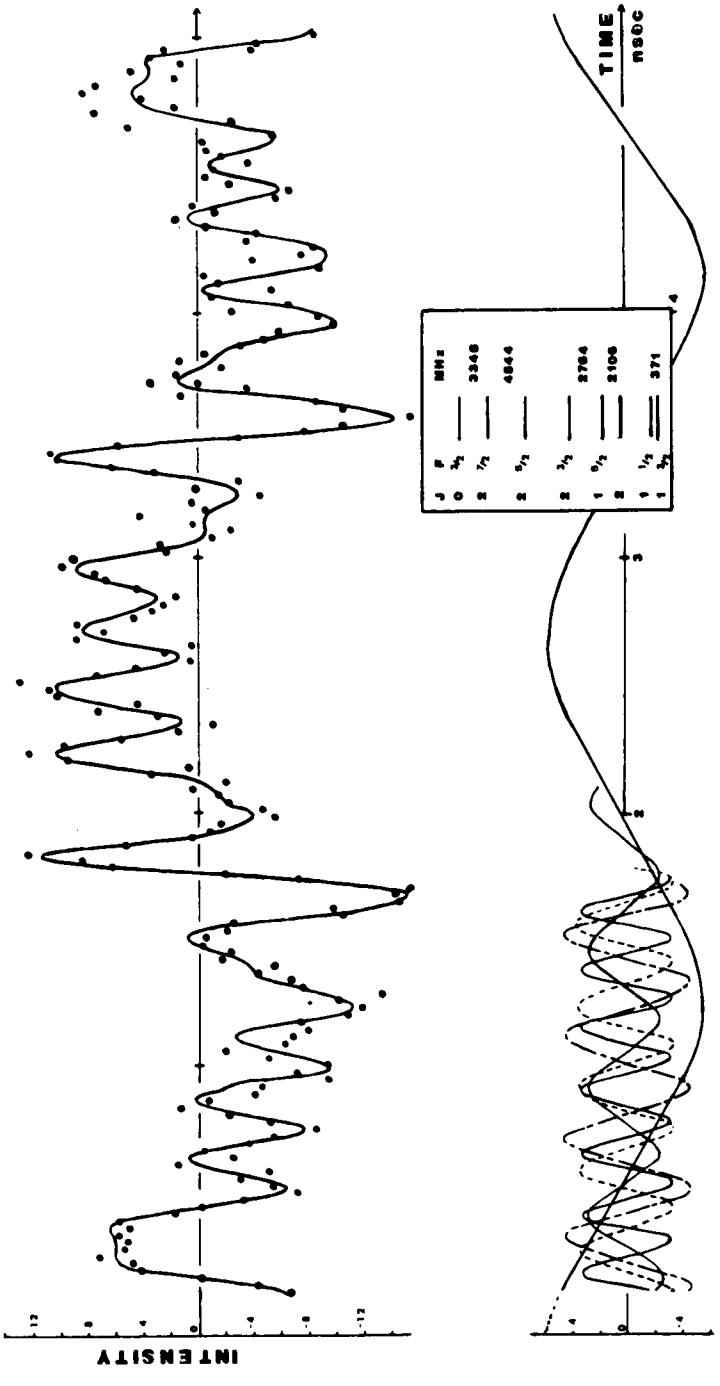

Fig.2. Zero field quantum beats of $3s\ ^3S - 4p\ ^3P$ in ^7Li II.

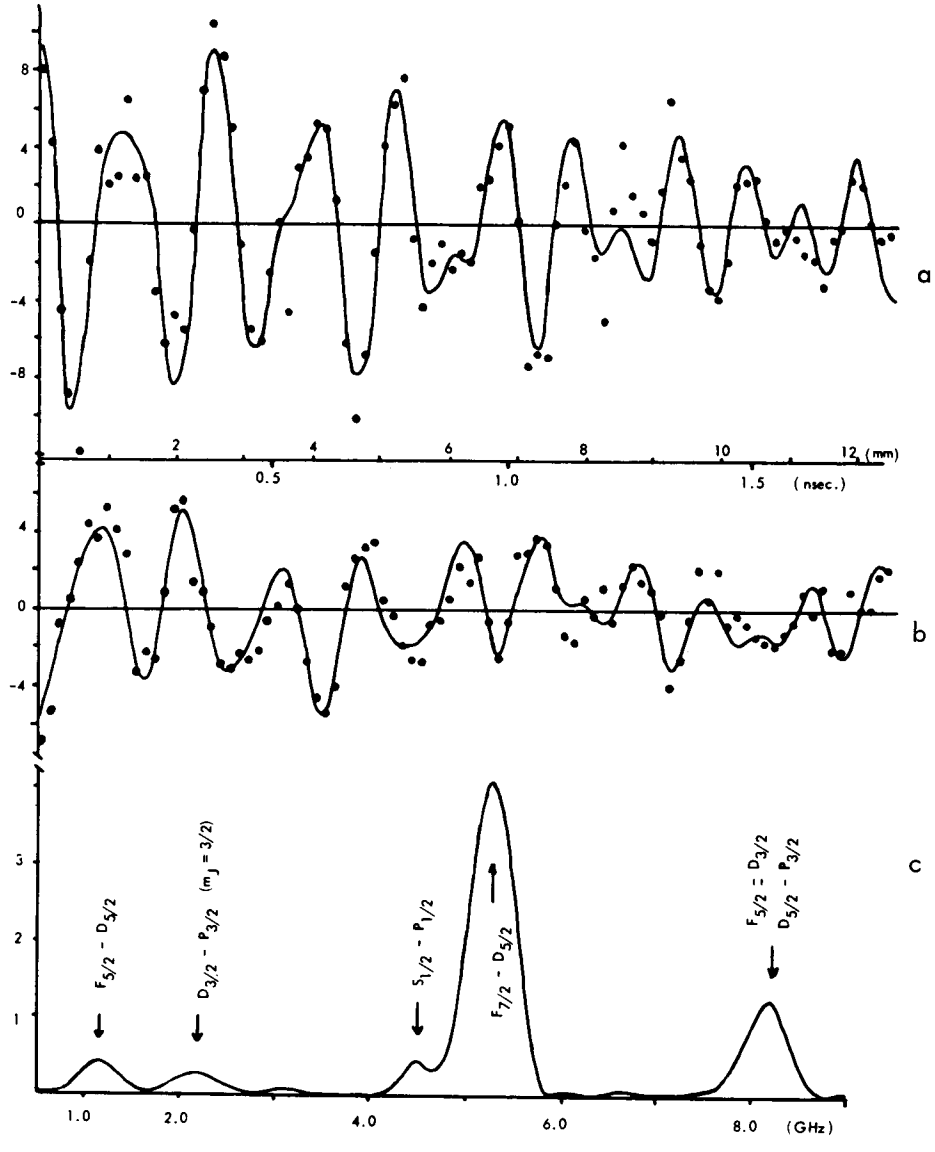

Fig. 3. Modulations in the decay of ^{4}He II n=3-4, 4686 Å in electric fields of $\pm$ 465 volts/cm in parts a and b (the solid lines are non-linear least-squares fits to sums of cosines), and the fourier transform of the difference curve in c. The modulation frequencies correspond to $m_J = \frac{1}{2}$ stark-shifted energy separations, except where noted.

superposition of these two states has been produced at the foil. Thus, the state is of non-definite parity with respect to reflections in the x-y plane, as well as not being an energy eigenstate.

Sellin et al.[8] and Gaupp et al.[9] verified this effect for n=2 of hydrogen, and we have shown that this is a general phenomenon[10] for hydrogenic states with observations of such "Eck-beats" in n=2,3 of HI and n=3,4 of ^{4}He II. In Fig. 3 we show an example of "Eck-beats" in ^{4}He II.

In the last part of Fig. 1 we indicate a further loss of source symmetry by tilting the foil so that its normal $\hat{n}$ is at an angle α to the beam-axis. An axial vector $\hat{n} \times \vec{v}$ can now be defined which corresponds to the possible production of atomic orientation, and consequently, circular polarized light may be emitted. Additionally, the atomic alignment becomes a three-component vector. Thus, a minimum of four parameters plus the population N are needed to describe each excited state. Farther source asymmetries need higher order state multipoles (see Refs. 4 and 5), but we shall here limit ourselves to the above examples where no external electromagnetic fields are disturbing the excitation process; such a description is then complete.

We should note that the last two examples assume that the beam-foil excitation depends on the final surface of the foil. Hence, these experiments are useful primarily to study this surface interaction. Hopefully, the isolation of surface interaction effects will also lead to information concerning the ionic states within the bulk of the foil.

2. The Tilted Foil Stokes Parameter Technique

In Fig. 4 we show the standard tilted foil geometry and define the relevant direction axes. We have previously shown[11] that we may describe an excited state produced in the foil interaction by a density matrix $\rho_{m\ m'}$, or ρ^k_q or the alignment vector $\mathcal{A}^c$ and orientation parameter O^c. The three sets of parameters are linearly related – m,m' are angular momentum projections, $k,q \leq |k|$ are irreducible tensor indices, and $\mathcal{A}^c$ and O^c are defined in Refs. 5 and 10.

Also, the light emitted from any excited state may be described completely in terms of the four Stokes parameters I,M,C and S. Thus, with respect to a set of axes ξ, η, ζ with ζ along the observation direction and, in our specific case, ξ denoting the "parallel" direction and η the "perpendicular" direction, the Stokes parameters are I, the total intensity, equal to the sum of the components of plane polarized light $I^{\parallel} + I^{\perp}$. M is the difference $I^{\parallel} - I^{\perp}$, while C is the difference in the two plane polarized components rotated at 45° to $I^{\parallel}$ and $I^{\perp}$, i.e., $C = I^{45°} - I^{135°}$. S denotes the net

circularly polarized light, $S = I_{r.h.} - I_{l.h.}$. The four parameters thus completely specify the polarization ellipse of the emitted light.[12]

The Stokes parameters are linearly related to all of the three sets of excited state parameters introduced above. Thus, for example,

$$M(\theta) = \sum_{k,q} a_{kq}(\theta) \cdot \rho^{k}_{q} \quad \cdots \cdot \qquad (2)$$

where the coupling coefficients a_{kq} depend upon the transition being observed and the angle of observation. The a_{kq} for a $^1P \rightarrow {}^1S$ transition are given in Ref. 10, and they can, in general, be derived from Refs. 4 or 5. The tensor component ρ^{0}_{0} describes the total number of excited atoms, while $\rho^{k=1}_{q}$ (or O^c) describes the atomic orientation, directly proportional to the circular polarization fraction S, and $\rho^{k=2}_{q}$ (or A^c) describes the atomic alignment. For electric dipole emission without external fields, only tensor components of $k \leq 2$ can be measured through this technique, while ρ itself may have undetermined tensor components of rank $k > 2$.

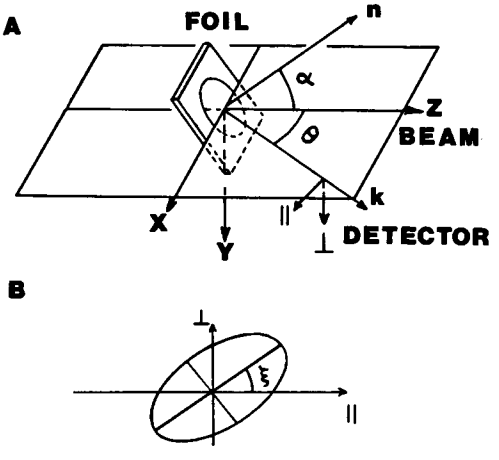

Fig. 4. A - Viewing geometry. The light vector k is in the $\hat{x}$-$\hat{z}$ plane, perpendicular to the $\hat{n}$-$\hat{y}$-$\hat{z}$ plane at an angle θ to the z axis.
B - The polarization ellipse.

The Stokes parameters are measured by observing the light at a particular angle θ through a fixed retardation plate and a rotating polarizer. Instrumental polarization can be eliminated by the introduction of a Hanle depolarizer[13] immediately after the polarizer. The retardation plate may be removed to measure the linear polarization parameters.

Thin carbon foils were either mounted in holders of various tilt angles, or mounted on a rotatable x-axis (see Fig. 4) which allowed a continuous variation of tilt angle α. The rotation of the polarizer was controlled by an on-line ASI computer in the Argonne experiments [14] or a motor drive control system in the Toledo experiments[14]. Light collected at each step was normalized either to Faraday cup current, or to a total light yield monitor observing only the transition of interest.

The first measurements using this Stokes parameter technique[11,14,15] indicated a very large surface effect in the beam-foil excitation. That is, the changes in M, C, and S with foil tilt angle α were large compared with their values at $\alpha=0$, and the asymmetric surface interaction produced circular polarization fractions of up to 25%.

3. Magnetic Field Quantum Beat Measurements of Asymmetry Parameters

For the cylindrically symmetric $\alpha=0$ beam foil source, the excited state may be aligned relative to the beam z-axis. Thus, a perpendicular magnetic field will induce a precession of twice the Larmor frequency ω_L of the classical damped electric-dipole oscillator as the excited ion moves downbeam. The theory of these magnetic field light intensity modulations has been discussed in detail by Gaillard et al.[16] for the case of cylindrical symmetry. It should be noted that a magnetic field parallel to the beam axis will produce no precession, and consequently no modulations.

When the cylindrical symmetry is broken by tilting the foil, a parallel magnetic field will induce modulations of frequency ω_L when the $q=1$ (for $k=1,2$) terms of the density matrix ρ^k_q are non-zero, while the perpendicular magnetic field will induce both $2\omega_L$ and ω_L modulations.

Church et al.[17,18] and Liu et al.[19] have observed such magnetic field modulations, and the phases and amplitudes of the modulations have been described in terms of the excited state parameters.[5,18] Hence, both the Stokes parameter measurements, and the magnetic field quantum beats lead to the same experimental results – the alignment and orientation parameters of the source.

4. The Structure of Unresolved Multiplets

The first verification of atomic alignment in the beam-foil source was the observation[3] of quantum beats from unresolved multiplet structures. We can expect that the other excited state asymmetry parameters can also be determined through similar observations using filted foil excitation. Ellis[4] has developed the general theory, and shown that: (1) atomic orientation, $\rho^{k=1}_q$, can be measured through observations of the time-modulation of the fractional circular polarization, (2) the atomic alignment, $\rho^{k=2}_q$, describes the time-modulations of the fractional linear polarization, and (3) the relative beat amplitudes, in the case of multiple frequencies, should remain unchanged, for a given type of polarization, as the foil tilt angle is varied.

We have verified[20] these results for the hyperfine structure quantum beats of $3s^3S - 3p^3P$ in ^{14}N IV, which are shown in Figs. 5 and 6.

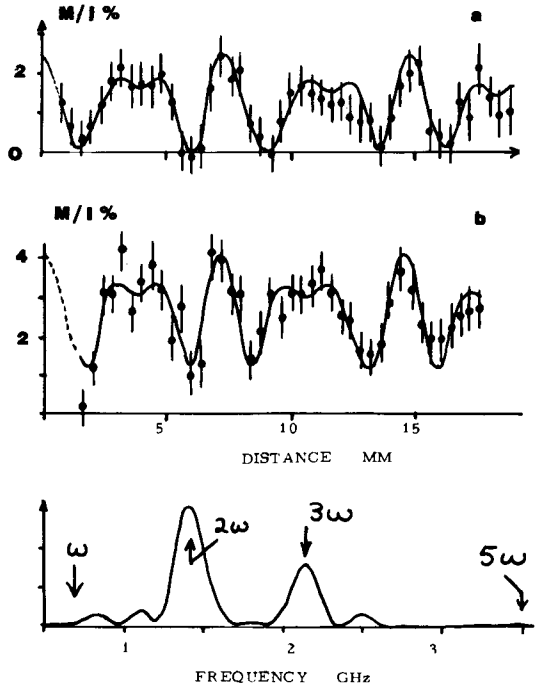

Fig. 5. Zero field quantum beats of ^{14}N IV $3s\ ^3S - 3p\ ^3P$, 3480 Å in linearly polarized light. a - 0° foil, b - 45° foil, c - fourier transform of b.

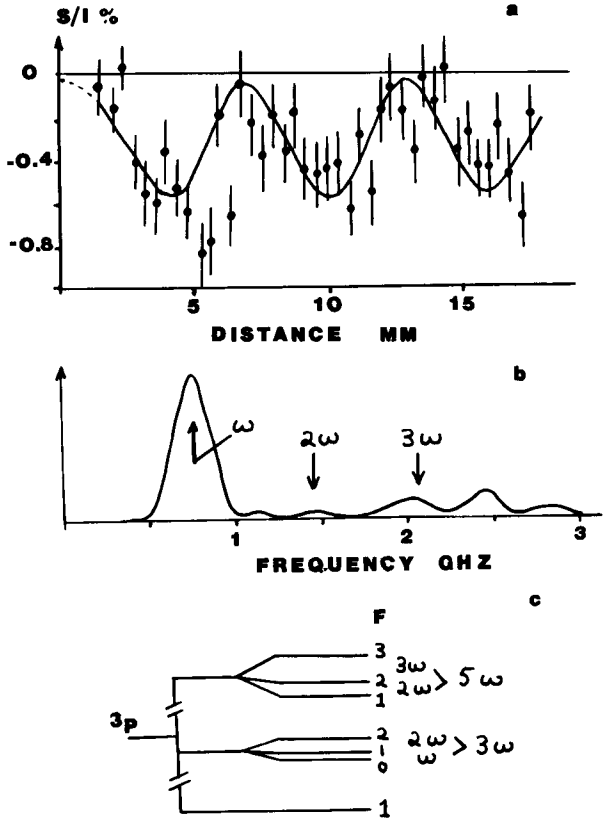

Fig. 6. a - zero field beats of ^{14}N IV 3s ^{3}S - 3p ^{3}P, 3480 Å, in circular polarized light, foil tilted at 45°, b - fourier transform, c - energy levels.

5. Theories of the Surface Interaction, and Comparisons with Experiment

Eck[21] has attempted to explain the initial results[11] of surface induced alignment and orientation by introducing an electric field perpendicular to the foil surface. This electric field removes the degeneracy in $|m_L|$ and transfers the alignment produced from excitation in the bulk into a coherence between states of different m_L. This is similar to the work of Lombardi[22] who has

shown that external electric fields skewed to an aligned excited state can produce orientation. However, Eck's theory, in particular for ^1P states, predicts that the total polarization $f_p = \sqrt{\{(M/I)^2+(S/I)^2+(C/I)^2\}}$ will be independent of foil tilt angle α. We have already shown[23] that f_p changes significantly with α and that Eck's simple theory must be modified.

Band[23] has included excited state production processes at the foil surface. He considers the moving ion to have an "active" electron which may be excited through interactions with the foil electrons and also by the surface potential barrier as it leaves the foil. It is essentially the interference between these two terms which gives rise to the orientation and alignment of the excited state. He obtains the following equations for the Stokes parameters of light emitted in a $^1S-{}^1P$ transition

$$\frac{S}{I} = -E \sin 2\alpha \sin\left(\frac{\omega}{v\cos\alpha}\right) \tag{3}$$

$$\frac{M}{I} = -E+F^2\cos 2\alpha+2E\sin^2 2\alpha \ \sin^2\left(\frac{\omega}{2v\cos\alpha}\right) \tag{4}$$

$$\frac{C}{I} = F^2\sin 2\alpha - E\sin 4\alpha \ \sin^2\left(\frac{\omega}{2v\cos\alpha}\right) \tag{5}$$

where E, F^2 and ω are constants of the surface, and v is the beam velocity. These equations are very similar to those of Eck,[21] but now the total polarization fraction f_p varies with α.

In Fig. 7, we compare Band's theory with our experimental data for $2s^1S-3p^1P$ in He I at 246 keV beam energy. The results show partial agreement but definite discrepancies appear.

Herman[24] has calculated the change in the perpendicular foil excitation matrix, $\rho(\alpha=0)$, for non-zero tilt angle α. He adds the contributions due to collisions between the moving ion and those surface atoms within its forward hemisphere as it leaves the surface. This introduces an addition due to the surface atoms on one side and a subtraction due to the lack of surface atoms on the other side of the moving ion. He then shows that the rank one density matrix components ($\rho^{k=1}$, 0^c, A^c_{1+}) should vary as tan α, while the rank two components ($\rho^{k=2}$ or A^c_{2+}) should remain fixed.

Thus, M/I should be constant, as is clearly not the case for the transition shown in Fig. 7. The k=1 component, S/I does not vary as strongly as tan α. However, our published results[23] for Ne III 2866 Å, $3s'^1D-3p'^1F$, do show a reasonable agreement for M/I and S/I.

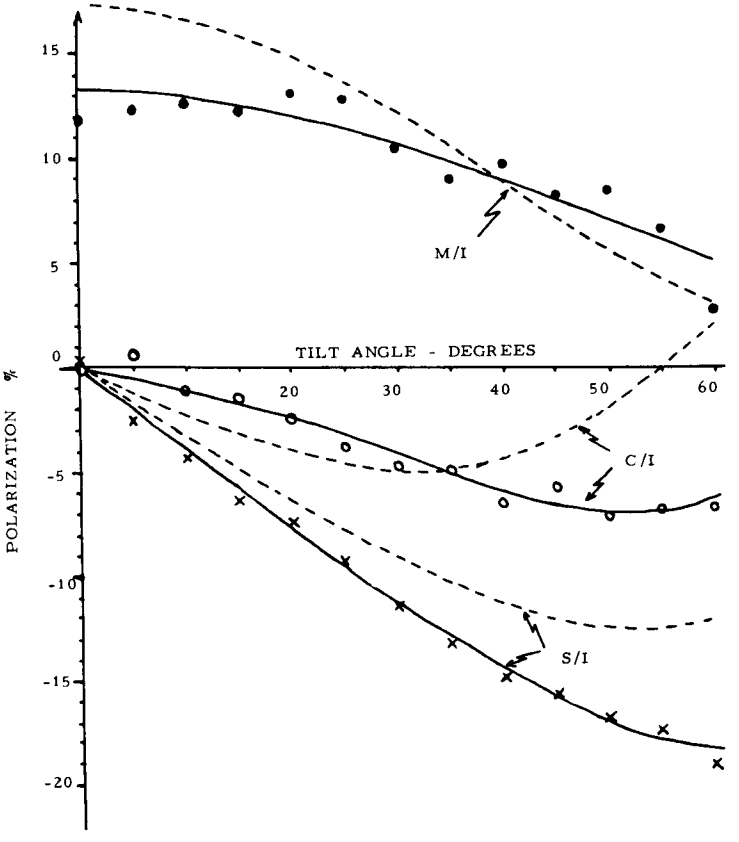

Fig. 7. Stokes parameters for ^{4}He I 2s ^{1}S - 3p ^{1}P, 5016 Å, ($\theta = 90°$, E = 246 keV) fitted to the Band theory - see Eqns. 3-5. The solid lines are independent fits to each Stokes parameter. The dashed lines are simultaneous fits with $E = -0.140$, $F^2 = +0.0328$, and $\omega/v = 0.73$.

The theories of Eck and Band assume that a state of well-defined parity is produced in the foil interaction. Consequently, the electric field interaction is a second order perturbation (present through the strong surface fields) and the Stokes parameters are functions of 2α, 4α, etc. A first order interaction should introduce terms proportional to α, 3α, etc., and we have noted earlier[20] that substitution of $\alpha/2$ for α would indeed give much better agreement with experiment.

Lombardi has pointed out that the production of non-definite parity states in the beam-foil process[26] allows such first order Stark effect processes to occur at the foil surface. He has derived expressions[27] for the Stokes parameters of a $^1S - {}^1P$ transition assuming s-p mixing. These can be expressed as expansions of $\alpha, 2\alpha, 3\alpha$, etc. Thus,

$$I = I_0 + I_1 \cos\alpha + I_2 \cos 2\alpha \tag{6}$$

$$M = M_0 + M_1 \cos\alpha + M_2 \cos 2\alpha + M_3 \cos 3\alpha + M_4 \cos 4\alpha \tag{7}$$

$$C = C_1 \sin\alpha + C_2 \sin 2\alpha + C_3 \sin 3\alpha + C_4 \sin 4\alpha \tag{8}$$

$$S = S_1 \sin\alpha + S_2 \sin 2\alpha \tag{9}$$

where $I_i (i=0\text{-}2)$, $M_i (i=0\text{-}4)$, $C_i (i=1\text{-}4)$, and $S_i (i=1,2)$ are functions of the various s and p density matrix elements. It should be noted that $M_i = C_i$ for all i except there is no C_0, and presumably p-d mixing, etc. would increase the number of terms in the expansions. For the case of no s-p mixing all α and 3α terms disappear, reproducing Eck's results with only a phase change at the surface, and Band's results on including surface excitation.

Unfortunately, this more general theory contains a large number of parameters (the many density matrix components) and also these parameters should vary with tilt angle α, since the surface interaction time changes as $1/v \cos\alpha$. This last variation is explicitly included in the result of Eck and Band – see Eqs. (3-5).

However, an analysis of our data for Ne III, 2866Å, in Fig. 8 shows an excellent fit to Lombardi's theory with only a small number of parameters for tilt angles between 0° and 80°. Less precise data for the ^{4}He I $2p\,{}^1P\text{-}4d\,{}^1D$ transition at 4922Å in Fig. 9 also show good agreement with the theory. The $1/v \cos\alpha$ dependence has been neglected, unlike the fit to Band's theory shown in Fig. 7.

A fit to the experimental data using the Lombardi theory gives estimates of various density matrix components. Thus, for the Ne III 2866 Å transition, we find $I_2 = M_4 = S_2 = 0$. These parameters are proportional to $\{\sigma(m = 0) - \sigma(m = 1)\}$ and hence indicate that $\sigma(m = 0) = \sigma(m = 1)$. However, we should than have $M_0 = 0$ which is certainly not true. An explanation may be that p-d mixing which should be as strong as s-p mixing has been neglected.

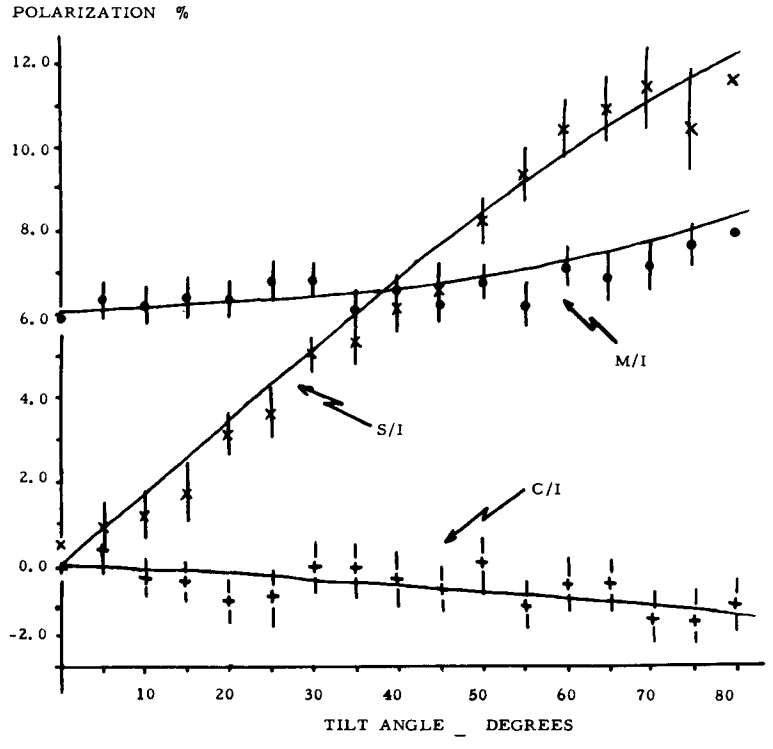

Fig.8. Stokes parameters for Ne III, 2866 Å, $3s'\,^1D - 3p'\,^1F$ ($\theta = 90^\circ$, E = 1 MeV), fitted to the Lombardi theory giving

$$I = 1 + 0.299 \cos\alpha$$
$$M = 0.090 - 0.012 \cos\alpha$$
$$C = -0.012 \sin\alpha$$
$$S = 0.130 \sin\alpha$$

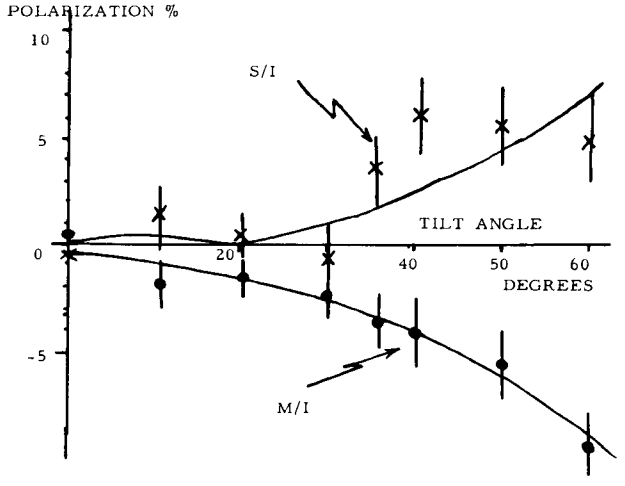

Fig. 9. Stokes parameters for ^{4}He I 2p ^{1}P - 4d ^{1}D, 4922 Å ($\theta = 90^\circ$, E = 325 keV), fitted to Lombardi parameters I = 1, M = -0.167 + 0.162 cosα C = 0, S = 0.16 sinα - 0.08 sin2α.

6. Conclusions

The theories of Eck, Band and Lombardi are all based on an interaction between a surface electric field and the moving ions. Their treatments differ in the types of excited states produced. The most general, that of Lombardi, seems to best agree with experiment in predicting the variations of the Stokes parameters with the foil tilt angle. Thus, within the limitations of only a few experimental data and the large number of fitted parameters of the theory, the surface interaction appears to be understood.

All experiments to date have taken place in relatively dirty vacuum conditions (about 10^{-6} torr), and detailed calculations of the types of excited states produced by particular foil materials will be useful when the experiments are repeated with clean surfaces in ultra-high vacuum. Such experiments are in progress.

We acknowledge with thanks the experimental help of Jean Desesquelles, Jerry Gabrielse, Tim Gay and Chuck Batson at Argonne National Laboratory and the University of Chicago. We had helpful discussions of the theories with Yehuda Band, Tom Eck, Roger Herman, and Murray Peshkin.

References

1. S. Datz, B. R. Appleton, J. R. Mowat, R. Lambert, R. S. Peterson, R. S. Thoe, and I. A. Sellin, Phys. Rev. Lett. 33, 733 (1974).

2. V. N. Neelavathi, R. H. Ritchie, and W. Brandt, Phys. Rev. Lett. 33, 302 (1974).

3. H. J. Andrä, Phys. Rev. Lett. 25, 325 (1970).

4. D. G. Ellis, J. Opt. Soc. Am. 63, 1232 (1973).

5. U. Fano and J. Macek, Rev. Mod. Phys. 45, 553 (1973).

6. I. C. Percival and M. J. Seaton, Phil. Trans. Roy. Soc. London A251, 113 (1958).

7. T. G. Eck, Phys. Rev. Lett. 31, 270 (1973).

8. I. A. Sellin, J. R. Mowat, R. S. Peterson, P. M. Griffin, R. Lambert, and H. H. Hazelton, Phys. Rev. Lett. 31, 1335 (1973).

9. A. Gaupp, H. J. Andrä, and J. Macek, Phys. Rev. Lett. 32, 268 (1974).

10. H. G. Berry, L. J. Curtis, D. G. Ellis, and R. M. Schectman, Proc. of the Stirling Conference on Electron and Photon Collisions with Atoms, July 1974, to be published, and R. M. Schectman et al., Proceedings of this conference.

11. H. G. Berry, L. J. Curtis, D. G. Ellis, and R. M. Schectman, Phys. Rev. Lett. 32, 751 (1974).

12. See, for example, D. Clarke and J.F. Grainger, Polarized Light and Optical Measurements (Pergamon Press, New York, 1971), Sec. 1.3.3. The Stokes parameters are defined in terms of the electric vectors in two arbitrary perpendicular transverse directions: $I = |E_{||}|^2 + |E_{\perp}|^2$, $M = |E_{||}|^2 - |E_{\perp}|^2$, $C = 2\,\mathrm{Re}(E_{||} E_{\perp}^*)$, $S = 2\ (E_{||} E_{\perp}^*)$.

13. W. Hanle, Z. Instrumentenk. 51, 488 (1931).

14. H. G. Berry, L. J. Curtis, and R. M. Schectman, Phys. Rev. Lett. 34, 509 (1975).

15. H. G. Berry, S. N. Bhardwaj, L. J. Curtis, and R. M. Schectman, Phys. Lett. 50A, 59 (1974).

16. M. Gaillard, M. Carré, H. G. Berry, and M. Lombardi, Nucl. Inst. Meths. 110, 273 (1973), and M. Gaillard, Thesè de Doctorat, 1974.

17. D. A. Church, W. Kolbe, M. C. Michel, and T. Hadeishi, Phys. Rev. Lett. 33, 565 (1974).

18. D. A. Church, M. C. Michel, and W. Kolbe, Phys. Rev. Lett. 34, 1140 (1975).

19. C. H. Liu, S. Bashkin, and D. A. Church, Phys. Rev. Lett. 33, 993 (1974).

20. H. G. Berry, L. J. Curtis, D. G. Ellis, and R. M. Schectman, Phys. Rev. Lett. 35, 274 (1975).

21. T. G. Eck, Phys. Rev. Lett. 33, 1055 (1974).

22. M. Lombardi and M. Giroud, C. R. Acad. Sc. (Paris) B266, 60 (1968), and M. Lombardi, J. Phys. 30, 631 (1969).

23. Y. Band, private communication, and Phys. Rev. Lett., to be published.

24. R. M. Herman, private communication, and Phys. Rev. Lett., to be published.

25. For example, 2s-2p excitation in hyrdrogen, Ref. 7-9, and in other hydrogenic states, Ref. 10.

26. M. Lombardi, private communication, and these conference proceedings.

Work supported in part by NSF and ERDA.