Role of Surface Interactions in Beam-Foil Excited-State Formation

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Polarization measurements have been performed on radiation emitted by He^+ ions (468.6 nm) in transmission (beam foil) and reflection (grazing incidence) geometries, with use of both carbon foil and silicon single-crystal targets. Comparison of the results of the two geometries and the two different targets indicates that the excited-state formation occurs at the exit surface. The impact of these results on current theories is discussed.

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Elliptic polarization of optical radiation observed in both grazing-incidence collisions[1,2] and tilted-foil transmission experiments[3] has received increasing attention stimulated by the suggestion of Fano and Macek[4] and Ellis[5] that the breakdown of cylindrical symmetry (by tilting the foil) could result in the production of an oriented nonstatistical magnetic sublevel population. The nature of the actual physical mechanism responsible for the phenomena remains a matter of controversy.[6,7] It has been argued that orientation develops either (a) from bulk-induced alignment transformed to orientation at the surface,[8-11] or (b) exclusively from the "last interaction" with the exit surface with no memory of the bulk.[12-14] Many workers have demonstrated experimentally that atomic orientation[1-3] depends strongly on the tilt (or grazing) angle relating the beam direction to the surface normal. While the above-mentioned experiments show conclusively that surface interactions are involved in the production of orientation, they do not make clear the possible role, if any, of prior polarization in the bulk.

In this Letter, we report on measurements of elliptic polarization from both ion-surface grazing-incidence collisions and tilted-foil ion-beam transmission experiments in order to elucidate the physical mechanism responsible for this phenomenon. We have compared the dependence of elliptic polarization on (1) bulk compositon (carbon and silicon targets), (2) bulk crystal orientation (channeling and nonchanneling transmission), and (3) collision geometry (tilted-foil transmission and grazing-incidence configurations). These are the first tilted-foil experiments involving a single-crystal target, and the first experiments comparing tilted-foil and grazing-incidence results with the same target. Previous measurements with single-crystal foils were performed at near-normal incidence only, and involved observations of light intensity and linear polarization in channeling and random directions.[15-17] Our observations lead us to suggest that there is no bulk contribution to the orientation and the same physical mechanism, namely a surface interaction, is responsible for both grazing-incidence and tilted-foil polarization phenomena.

The experiments were performed on the Bell Laboratories 2-MeV Van de Graaff accelerator, which supplied a He^+ ion beam into a target chamber at a vacuum of $\sim 1\times10^{-7}$ Torr. The target area was surrounded by a metal enclosure maintained at liquid-nitrogen temperature. The He^+ beam was varied in energy to compensate for tilt-angle–dependent energy loss, such that the emerging energy was maintained at 530 keV. Polarization data were acquired with a 0.3-m monochromator, a Polaroid HN-38 polarizer, a depolarizer which removes spectrometer polarization bias, and a retardation plate with one quarter-wave retardation at 468.6 nm. Single-photon counting techniques were used to detect and process photons emitted normal to the plane defined by the incoming beam and surface normal for both tilted-foil and grazing-incidence configurations as shown in Fig. 1. Crystal orientation was monitored by Rutherford backscattering.

The optical radiation observed in this experiment arises from the decay of the $n=4$ to $n=3$ states of hydrogenic He^+. For the tilted-foil experiments both polycrystalline carbon foils and a 0.5-μm (110) single-crystal silicon foil were

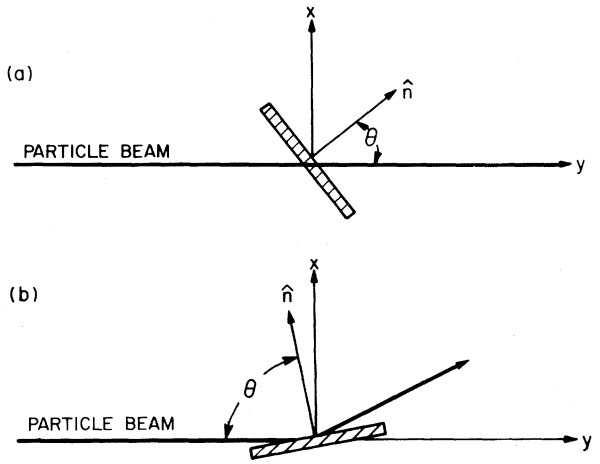

FIG. 1. Schematic illustrations of the experimental configuration: (a) Tilted-foil case. (b) Grazing-incidence case. Photons emitted in the $-z$ direction (out of the page) are detected.

used in the configuration shown in Fig. 1. The tilt angle θ, defined as the angle between the surface normal and the beam direction, was varied from $-80°$ to $+80°$. The measured polarization is expressed in terms of the three normalized Stokes parameters defined in the following standard way[18]:

$$S/I = (I_{RHC} - I_{LHC})/(I_{RHC} + I_{LHC}),$$
$$M/I = (I_0 - I_{90})/(I_0 + I_{90}), \quad (1)$$
$$C/I = (I_{45} - I_{135})/(I_{45} + I_{135}),$$

where I_{RHC} and I_{LHC} are the intensities of right-handed and left-handed circular polarization, and I_0, I_{45}, I_{90}, and I_{135} are the intensities of radiation with direction of polarization 0°, 45°, 90°, and 135°. 0° is along the x direction and 90° along the y direction as shown in Fig. 1. For this transition, circular polarization arises from an oriented initial state, i.e., a state with nonzero projection of angular momentum along the z axis. Linear polarization reflects alignment of the initial states.

The absolute magnitude of S/I is seen to rise monotonically with angle (Fig. 2). As shown, the transmission measurements for carbon films and single-crystal silicon provided almost identical results. The behavior of S/I is found to be independent of (a) bulk material and (b) crystal orientation. Passage through a number of unspecified channel directions, where the intensity of backscattering particles was observed to decrease up to a factor of 5, resulted in no alteration in the magnitude of S/I as shown in the inset in Fig. 2. In all cases C/I remained less than 2%. In addition, M/I was also small and was measured to be 2% or 3% for all angles. The polarization is found, therefore, to be almost entirely circular.

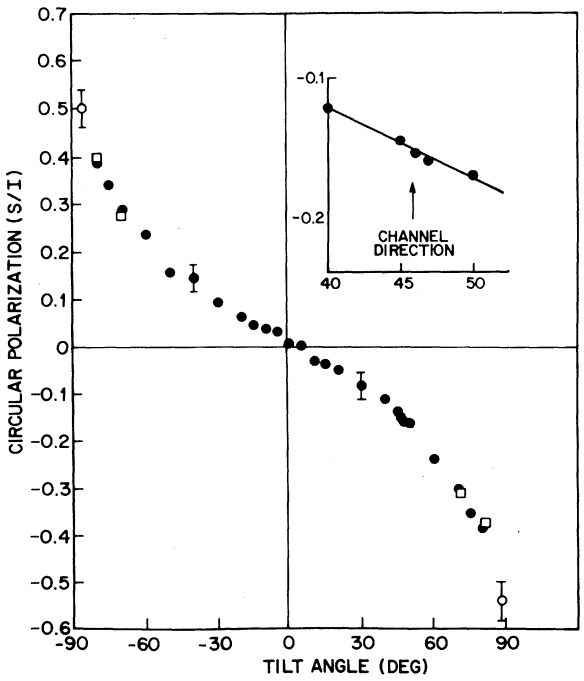

FIG. 2. Circular polarization, S/I, of He^+ ($n = 4$ to $n = 3$) at 468.6 nm as a function of tilt angle θ at a constant emerging energy of 530 keV for both grazing incidence with a silicon crystal (open circles), and for beam-foil transmission with a silicon crystal (solid circles), and a carbon foil (open squares). The inset shows the tilt-angle dependence of S/I in the vicinity of a low-index channeling direction with channeling critical angle of approximately 0.5°. The channeling direction was indicated by a factor of 5 decrease in backscattered yield.

The normalized Stokes parameters S/I, M/I, and C/I arising from grazing incidence collisions were also measured on the same surface of the silicon single-crystal target from which the particles emerged in the tilted-foil case. The experiments were performed at a grazing angle of approximately 4°. In these experiments, the normalized Stokes parameters expressing linear polarization, M/I and C/I, were also found to be small, less than 2%. However as shown in Fig. 2, the circular polarization, S/I, was found to be large, exceeding 50%. It should be noted also that the grazing-incidence measurements appear to smoothly join to the tilted-foil trans-

mission measurements when plotted on a scale which relates the surface-normal direction to the beam direction (see Fig. 2). This observation suggests that the tilted-foil and grazing-incidence polarization phenomena are closely related.

The present experiments provide convincing evidence that (1) the mechanism responsible for the polarization at large tilt angles is the same for both the tilted-foil and the grazing-incidence cases and (2) that the creation of excited states occurs at the surface with no memory of interactions in the bulk. Our conclusion that the polarization is entirely due to a surface interaction is supported by the following observations: (a) The orientation is measured to be independent of bulk material, carbon or silicon. For these experiments performed at only moderate vacuum (10^{-7} Torr) the surface composition is unlikely to be entirely bulk related and both targets are likely to be similarly contaminated. Thus a surface mechanism could account for the identical polarization results observed for carbon and silicon. (b) The observed polarization was found to be independent of low-index channel directions of transmission through the silicon crystal (see inset in Fig. 2). Note than in an amorphous solid, the coherence of any excitation produced in the bulk must exhibit axial symmetry about the beam direction. Thus alignment (M/I) can be produced, but in the absence of a surface interaction orientation is forbidden. Note also that this symmetry constraint no longer applies to a single-crystal sample; i.e., the creation of orientation in a crystalline solid is no longer ruled out.[19] Our observation that S/I is independent of low-index channel direction demonstrates that the possible production of orientation in the bulk does not contribute significantly to the measured orientation. (c) It has been established by computer simulation[2] that the vast majority of grazing-incidence projectiles at keV energies that are scattered near the specular angle do not penetrate deeper than 2 or 3 atomic layers. Thus the grazing-incidence mechanism is certainly dominated by a surface interaction. Our observation that the grazing-incidence results join smoothly with the large-angle transmission results suggests a similar mechanism for the latter.

We conclude from these arguments that the coherent superposition of excited states which gives rise to the emission of polarized light has no memory of bulk interactions. This is in contradiction to the viewpoint[8-11] that an axially symmetric coherence is produced in the bulk, and that the role of the surface interaction is merely to convert partially the coherence into orientation. We note that Eck's original model[8] requires that the total polarization, $(M^2+C^2+S^2)^{1/2}/I$, remain constant as a function of tilt angle θ. This is in drastic disagreement with our results. However, extensions of the model which incorporate coherence among states of different orbital angular momentum L are no longer subject to this constraint.[9-11] Band[11] has calculated the evolution of the most general axially symmetric density operator to second order in $\Delta = \int V(r_\perp) dr_\perp/\hbar v_\perp$, where $r_\perp$ is the distance normal to the surface, $V(r_\perp)$ is the energy splitting of atomic levels near the surface, and $v_\perp$ is the component of particle velocity normal to the surface. He obtains expressions for the Stokes parameters involving five unknown constants which can be adjusted to best fit experiment. Our data can be fit closely by these expressions for $\theta < 60°$, but cannot be fitted well at larger angles. Unfortunately, this fit does not allow us to make any conclusions about the mechanism of excited-state production. If Band's analysis were carried out to infinite order in Δ, then any variations of the Stokes parameters consistent with the symmetry of the experiment could result. Success or lack of success in fitting experiments with the second-order expressions does not reflect at all on the validity of the underlying physical picture.

These experimental results provide compelling evidence that the excited states are formed at the exit surface, not in the bulk. The precise mechanism of formation has not been established. Oriented and aligned excited states could be produced by electron pickup as the ions recede from the surface. This mechanism is consistent with all grazing-incidence and transmission results reported to date.[12-14,20] However, contributions from other surface mechanisms such as selective deexcitation,[20] anisotropic atomiclike collisions,[21] and plasmon scattering[22] cannot be excluded at this time.

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[19]Orientation can arise in principle in situations where reflection symmetry about the beam direction is absent. This could occur, for example, for beam transmission in any but a few low-index directions throught a crystal, or in the presence of nonrandom electric or magnetic fields in any solid.
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Observation of Competition of Rotational Effects in the Intensity of Ultraviolet Bands of OH

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The band oscillator strength of OH was found to be essentially independent of rotational excitation for the (1,0) transitions, but to decrease with rotation for the (0,0) transitions. This difference in the rotational dependence is a result of competition between a rotationally induced decrease in the electronic transition moment and rotationally induced changes, which may be either an increase or decrease, in the Franck-Condon factor for these bands.

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The band intensity of rotational-electronic transitions in the ultraviolet bands of OH has been known to vary with rotational excitation.[1] In the case of the (0,0) transitions, this variation is largely a manifestation of the fact that the electronic transition moment of OH decreases with increasing internuclear spacing and that this latter spacing is lengthened in the presence of rotation. Although the Franck-Condon factor for the (0,0) transitions also decreases with rotational excitation, this decrease is much smaller than that associated with the electronic transition moment,[2] and was often neglected in many previous discussions.[3,4] The purpose of this Letter is to report the first observation that the band oscillator strength for the (1,0) transitions is, unlike that for the (0,0) transitions, essentially independent of rotational excitation. This observation dictates that rotational stretching of the internuclear spacing leads to a decrease in the electronic transition moment and an almost equal amount of increase in the Franck-Condon factor