be aligned in the process. If alignment were the answer to the problem, one would surely expect to observe beats on the decay curve associated with the $3^2P_{3/2}^\circ$ level, which can be aligned, but they were found to be absent.

If the $3^2P_{1/2}^\circ$ level is neither aligned or oriented, then one must look for mechanisms that would modulate the total intensity of the radiation. It is quite conceivable that stray fields may Stark-mix levels of high n and l of opposite parity (for such hydrogenic levels, only very small fields would be necessary) and that the total intensity of the transition out of the $3^2P_{1/2}^\circ$ is modulated by the transfer of the total-intensity modulations of these high-n levels in the process of cascading. The apparent presence of oscillations of approximately the same frequency on the decay curve of the feeding transition, $3^2P_{1/2}^\circ$-$3^2D_{3/2}$, would tend to support this idea of cascade transfer. In addition, the persistence of the oscillations to regions of the decay curve dominated by the cascade contributions from long-lived "yrast" levels again supports the hypothesis that the observed phenomenon may be associated with the cascade transfer of Stark-induced total-intensity modulations of the long-lived "yrast" levels of high n which are "mixed" by small stray fields. If this is indeed the case, it is rather surprising that the modulations are not averaged out by the large number of different frequencies that is to be expected in such a situation. It is also puzzling why this cascade-transfer process should selectively modulate the $3^2P_{1/2}^\circ$ level and not the $3^2P_{3/2}^\circ$ level.

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Spatial Distribution of Orientation of Fast Ions Excited by Surface-Grazing Collisions*

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We have investigated the grazing-angle scattering of fast ions from solid surfaces. We establish that the circular polarization of light emitted from the scattered ions, and hence the ion orientation, varies strongly with the ion scattering angle. We show that the nearly elastic ion-surface collisions produce maximum allowed orientation of states with very small alignment.

An important recent development in accelerator-based atomic physics is the production of excited-ion beams with a significant degree of orientation. This has been accomplished both by directing ions through a thin tilted foil,[1] and by scattering ions at a grazing angle from a solid surface.[2,3] The large induced orientation may be useful in studying atomic-structure parameters of ions,[3] in nuclear-structure measurements,[4] and also in studying surface structure and the ion-beam–solid collision process itself. In this Letter, we report the first measurements of the dependence of the observed orientation on the angle of scattering of beam ions from a surface. A strong angular dependence is found which both clarifies the interaction mechanism and points to the possibility of quantitative *measurements of* atomic- and nuclear-structure parameters.

Recent measurements by Andrä and co-workers[2,3] have shown the existence of large orientations of Ar II excited states, when excited by grazing collision with solid Cu surfaces. They suggested from measurements of angular distributions of ion currents that at incidence angles of less than 5° the Ar^+ ions were scattered mainly in the specular direction.

The purpose of this Letter is to show that the orientation of each excited state depends sensitively on the angle of scattering. We have measured the spatial distributions relative to the excitation surface of the light yield, and of the three relative Stokes parameters[1] M/I, C/I, and S/I describing the alignment and orientation of the excited states. Our observations clarify the angular-momentum characteristics of the ion-surface interaction, explain the difficulty of making Hanle effect and high-field level-crossing measurements,[3] and suggest how to make such techniques quantitative for fine and hyperfine structures in heavy ions.

Figure 1(a) shows the experimental arrangement. The target was electrically isolated at +300 V and the net collected current was used as a normalizing signal. The tilt-angle drive screw (Fig. 1) was adjustable for varying the angle of grazing incidence α, and the entire assembly of the tilted surface and second collimator could be moved parallel to the beam axis. A single lens focused the light emitted at 90° (in a solid angle of ~0.09 sr) through a polarization-analysis system and into the monochromator.

The observation region sampled by the detector is an image 1.5 mm high by 0.2 mm wide as shown in Fig. 1(b). This observation region could be moved both along the beam (as shown) and positioned at different heights above the incident-beam axis. The polarization-analysis system allows simultaneous measurement[5] of the four Stokes parameters I, M/I, C/I, and S/I, where the total intensity I has been normalized to the net beam charge. The targets consisted of solid blocks of polished copper or gold. Following cleaning with a 30-μA Ar^+ (1.0 MeV) beam, our results were very reproducible. Similar results were obtained with both surfaces, but we shall discuss here only the more detailed results for copper. The background pressure was 5×10^{-7} Torr with pumping from a liquid-nitrogen–trapped oil diffusion pump.

Two-dimensional spatial mappings of the four Stokes parameters were obtained by successive measurements at steps along the beam axis for given heights of the observation region above the excitation surface Fig. 1(b). A partial set of these measurements is shown in Fig. 2 for the total intensity I and the circular-polarization fraction S/I for the Ar II transition $4s^1\,{}^2D_{5/2}$-$4p^1\,{}^2F_{7/2}$. Excitation in this case was by a copper surface at a tilt angle $\alpha = 5.5°$ relative to the incoming 1.0-MeV Ar^+ ions. The data scatter and reproducibility indicate that the accuracy is close to the statistical accuracy of about 1%. Systematic errors due to surface variations, beam-current normalization, and detection nonlinearities,

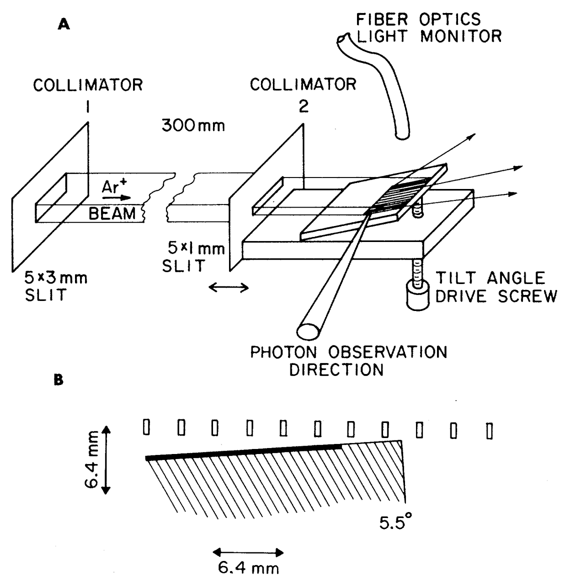

FIG. 1. (a) shows the experimental arrangement. (b) indicates the positioning of the surface relative to the rectangular observation areas (0.2×1.5 mm^2) from which light was observed. A series of positions along the beam is shown at a fixed height above the surface. The height was also adjustable.

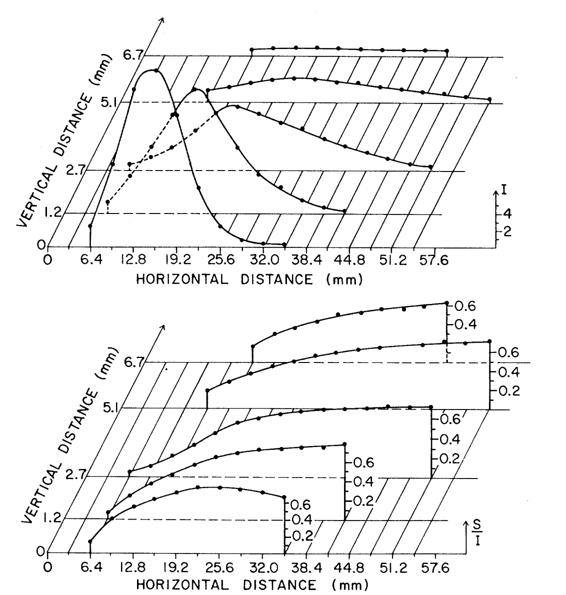

FIG. 2. Total intensity (top), and circular polarization fraction S/I (bottom) of Ar II 4610 Å shown as two-dimensional functions of the horizontal distance along the beam and the vertical distance above the surface. The interaction region is located between 0 and 19.2 mm, is tilted at 5.5° to the horizontal and its top-most point is near zero in the vertical direction.

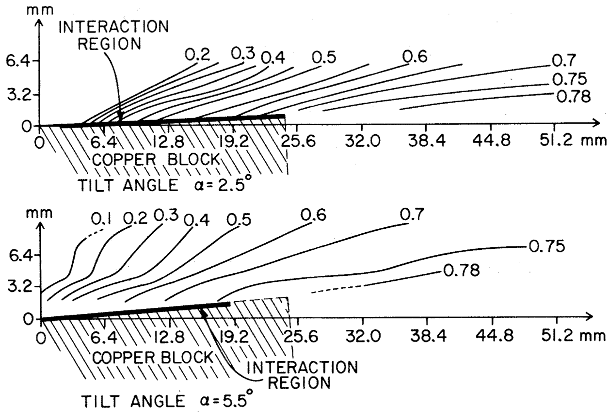

FIG. 3. Contours of equal circular polarization S/I for Ar II, 4610 Å for copper surface tilt angles of 2.5° and 5.5°.

are expected to be less than 5%.

We have converted the two-dimensional sets of circular-polarization measurements to a series of contour plots. Thus Fig. 3 shows the contours of constant S/I where the two coordinates are the distance along the beam axis and the distance above the surface. The maximum value observed for S/I is the same (0.78) for both incident grazing angles of $\alpha = 2.5°$ and 5.5° Observations made very near the surface are complicated by lifetime effects, velocity distributions, and averaging over a range of scattering angles. However, we can conclude from the S/I contours that this maximum circular polarization comes only from particles scattered close to the specular-reflection angle. S/I is more forward peaked for the smaller grazing angle of 2.5° in Fig. 3, in agreement with this conclusion. Ions scattered through larger angles (and in this same excited state) give lower values of S/I, and hence are less oriented. The linear-polarization fractions M/I and C/I (referred to the beam direction) were very small compared with S/I for surface tilt angles up to 12°, for Ar^+ beam energies between 0.5 and 2.5 MeV and for some fifteen Ar II transitions measured. M/I and C/I were generally less than 0.08 and 0.03, respectively.

Our optical measurements enable us to determine the spatial distributions of definite excited states. We observe that the circular-polarization fraction is greatest from the forward-scattered ions which have undergone nearly elastic scattering with the surface. Their net orientation comes either at a single momentum-transfer collision or during their relatively slow motion away from the surface. Much less circular polarization is observed from ions undergoing large scattering angles which may have penetrated into the surface. For the larger surface tilt angles, the forward-scattered-particle fraction is reduced relative to that for large-angle scattering. This can explain the two low values of S/I obtained by Andrä *et al.*[3] for tilt angles of 5° and 10°, whereas we find that the forward-scattered ions show strong circular polarization for tilt angles up to 12°.

The statements that we make relating the atomic properties (alignment and orientation) to the measured photon properties (the Stokes parameters) are based on the following equations:

$$
\begin{aligned}
M/I &= [a/(1+A)]\langle L_x^2 - L_y^2\rangle/\langle L^2\rangle,\\
C/I &= [a/(1+A)]\langle L_xL_y + L_yL_x\rangle/\langle L^2\rangle, \qquad (1)\\
S/I &= [-b/(1+A)]\langle L_z\rangle/\sqrt{\langle L^2\rangle},
\end{aligned}
$$

where z is the observation axis, and x is the beam axis. The constants a and b depend on the coupling scheme and on the transition involved.[6] For the Ar II transition $4s'\,^2D_{5/2}$-$4p'\,^2F_{7/2}$, $a = \frac{15}{14}$ and $b = 27\sqrt{3}/28$. The correction to the denominator is

$$A = a(\tfrac{1}{3} - \langle L_z^2\rangle/\langle L^2\rangle), \qquad (2)$$

which is small for a small electric quadrupole moment $\langle 3L_z^2 - L^2\rangle$.

A surprising feature of these measurements for the forward-scattered-ion fraction is the very strong orientation (S/I) and almost zero alignment (M/I and C/I). These results show that the quadrupole moments are small, and the excitation is nearly symmetric about the observation axis. This is perpendicular to the beam axis, which is the usual axis of symmetry in beam-type collisions. Thus, we are led to consider the limiting case of a state of *maximum orientation with no alignment.* If we choose the observation direction as the z axis, then by the symmetry of the collision, $\langle L_x\rangle = \langle L_y\rangle = 0$ and for no alignment $\langle L_x^2\rangle = \langle L_y^2\rangle = \langle L_z^2\rangle = \frac{1}{3}\langle L^2\rangle$. By requiring only that excitation probabilities be positive, we obtain

$$\langle L_z\rangle = \tfrac{1}{3}(L+1). \qquad (3)$$

Applying this to an LS-coupled state (SLJ) and assuming a spin-independent interaction, we find that the circular-polarization fraction is

$$\frac{S}{I}=\left(\frac{L+1}{2L}\right)^{1/2}\frac{\begin{Bmatrix}L & L & 1\\ J & J & S\end{Bmatrix}\begin{Bmatrix}1 & 1 & 1\\ J & J & J'\end{Bmatrix}}{\begin{Bmatrix}L & L & 0\\ J & J & S\end{Bmatrix}\begin{Bmatrix}1 & 1 & 0\\ J & J & J'\end{Bmatrix}}, \tag{4}$$

for the transition to a final state J'.

For the Ar II, 4610-Å transition Eq. (4) gives $S/I=0.64$, which is close to, but very definitely less than, our measured maximum for S/I of 0.78 ± 0.02. However, we should note that measurements of M/I and C/I yield only two of the three nonzero alignment parameters. The alignment term $\langle 3L_z^2-L^2\rangle$ has not been measured, but it is straightforward to show that only small amounts of this term are necessary to make our observed circular-polarization fraction agree with this limit. Other Ar II transitions (e.g., as shown in Ref. 3) give similar results of low M/I and C/I, and S/I close to the limit given by Eq. (4). These results suggest that the ions which we have measured have approximately the maximum orientation possible, given a small amount of alignment. Although spectral blending causes low precision in may cases, the orientation parameter $\langle L_z\rangle/(\langle L^2\rangle)^{1/2}=\langle L_z\rangle/[L(L+1]^{1/2}$ is approximately constant.[7]

In conclusion, we have established that ion orientation is strongly dependent upon scattering angle in grazing collisions with metallic surfaces. We show that nearly elastic collisions produce ions which have the maximum orientation for a small alignment. Less-elastic collisions produce less-oriented ions. Quantitative Hanle effect and level-crossing measurements of oriented ions will therefore be possible if the forward-scattered component alone is observed. The produced orientation is in the same direction for all observed transitions. This is important in experiments to determine the signs of nuclear g factors using the hyperfine interaction to couple to the surface-collision-induced electronic polarization.[4]

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