Orientation of Atoms by Collisions with Inclined Surfaces

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We have measured large orientations for Ar II states excited by 300-keV Ar^+ impact on solid Cu surfaces at grazing incidence. We report circulary polarized light fractions of up to 76% and interpret the excitation mechanism as an orbital-angular-momentum effect. As an application of this new excitation scheme, a zero-field level-crossing experiment is presented.

We report new experiments which confirm and extend the recent observation that the excitation of fast ion beams by impact with inclined solid amorphous surfaces can lead to appreciable excited-state orientations.[1] We present measurements of angular distributions of the scattered ion current which support the assumption that near grazing incidence, there is a well-defined direction for the outgoing oriented atoms, as in "tilted-foil" experiments.[2] We also give the first results for the application of this "tilted-surface" effect to atomic level-crossing studies.

The experimental setup is shown in Fig. 1. With the axes indicated, the direction of observation was along Ox. The incident beam direction is along Oz, the target surface normal makes an angle α with the $-y$ axis in the yz plane, and we anticipate the results given below by ascribing an angle θ to the average forward-scattered particle direction. The target was amorphous Cu, of dimensions 30 mm×15 mm×1 mm, and its surface was mechanically polished flat to 2 μm. It was mounted on a shaft which was along Ox, and the angle α could be set to a precision of $\pm 0.17°$. A 1-mm-diam pickup wire was mounted parallel to and off center from the x axis in such a way as to describe a circle of radius 7 cm about that axis. The scattered beam current could be measured with this wire as a function of γ, in 1.4° steps. The pickup wire and the target could be held independently at variable potentials to suppress secondary-electron emission.

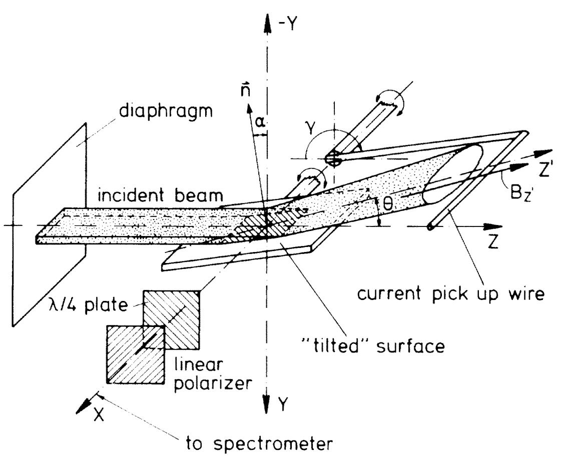

FIG. 1. Experimental setup. The beam cross section is $0.5\times 5\ \mathrm{mm}^2$.

The optical system consisted of an achromatic $\lambda/4$ plate and a linear polarizer, followed by a single imaging lens, a 0.3-m grating spectrometer, and a photomultiplier tube in photon-counting mode. The viewing region was approximately 7 mm in diameter in the yz plane, and was centered on the x axis. Spectra were built up by repetitive sweeping of the wavelength proportional to collected charge of the unbiased target (our results are independent of secondary-electron emission) and recording the photon counts in a synchronized multichannel scaler. Similarly level-crossing curves were obtained by sweeping the magnetic flux $B_{z'}$ proportional to time at less than ± 5% beam fluctuations and recording photons of a particular line and polarization. $B_{z'}$ was applied by Helmholtz coils aligned along the scattered beam direction Oz'.

In order to determine excited-state orientations of free scattered ions we measured the normalized Stokes parameter $S/I = (I\sigma^- - I\sigma^+)/(I\sigma^- + I\sigma^+)$ by rotation of the $\lambda/4$ plate in steps of 90°, where $I\sigma^-$ and $I\sigma^+$ are, respectively, right and left circularly polarized light intensity in optical convention.[3] Values of S/I for 17 Ar II lines excited by 300-keV Ar^+ impact on Cu targets at $\alpha = 1.5°$ are given in Table I. They belong to five multiplets[4] and were taken from two spectra recorded with

TABLE I. Measured circular polarization fractions $(S/I)_{\rm exp}$ at $\alpha = 1.5°$ and comparison with theoretical values $(S/I)_{\rm theor}$.

Multiplet	J_0 lower state	J upper state	Wavelength [Å]	S/I exp	S/I theor
$4p\,^2D°$	3/2	5/2	4879.9	0.65(4)	0.68
↓	3/2	3/2	4726.7	0.33(5)	0.31
$4s\,^2P$	1/2	3/2	4965.1	0.64(4)	0.72
$4p'\,^2F°$	5/2	7/2	4609.6	0.76(4)	0.66
↓	5/2	5/2	4637.2	0.17(7)	0.21
$4s'\,^2D$	3/2	5/2	4589.9	0.72(4)	0.69
	5/2	7/2	4348.1	0.62(4)	0.63
$4p\,^4D°$	5/2	5/2	4266.5	0.17(4)	0.16
	3/2	5/2	4426.0	0.60(10)	0.55
↓	3/2	3/2	4331.2	0.21(3)	0.20
$4s\,^4P$	1/2	3/2	4430.2	0.57(10)	0.50
	1/2	1/2	4379.7	0.43(10)	0.50
$4p\,^4P°$	5/2	5/2	4806.0	0.15(3)	0.18
↓	5/2	3/2	4735.9	−0.14(4)	−0.17
	3/2	5/2	5009.3	0.50(10)	0.60
$4s\,^4P$	3/2	1/2	4847.9	0.10(3)	0.14
$4p\,^4P°$ ↓ $3d\,^4D$	7/2	5/2	4401.0	−0.27(10)	−0.43

$\Delta\lambda = 6$ Å full width at half-maximum, running from 4000 to 5500 Å in σ^+ and σ^- light, respectively, showing clear Ar II line spectra including only some strong Cu I lines and weak Cu II lines from sputtered Cu. The errors quoted are composed of statistical counting and spectral blending uncertainties but do not account for surface properties. These are determined by the degree of polish prior to installation, the deposition of impurities under ion impact,[5] and cleaning by sputtering.[6] The latter two are competing effects which, in measurements at 2×10^{-6} Torr with low beam flux ($\sim 1\ \mu$A/0.5×5 mm^2) at 1°, led to impurity deposition and low values of $S/I \simeq 0.34$ for the 4609-Å line as in Fig. 2. For the results in Table I, the Cu target was exposed to a beam flux $\sim 9\ \mu$A/2.5 mm^2 and appeared as a clean and highly polished mirror thereafter.

As a first stage in the understanding of the anisotropic excitation process, it is of value to determine the angular distribution of the scattered ions. The difficulty of applying available scattering results from the literature[7] to our conditions (2×10^{-6} Torr, 1 μA/2.5 mm^2, surface granulation $\sim\mu$m) led us to investigate the scattering of He^+, Ar^+, and Sn^+ beams at various energies and angles α with our target held at +280 V. Typical

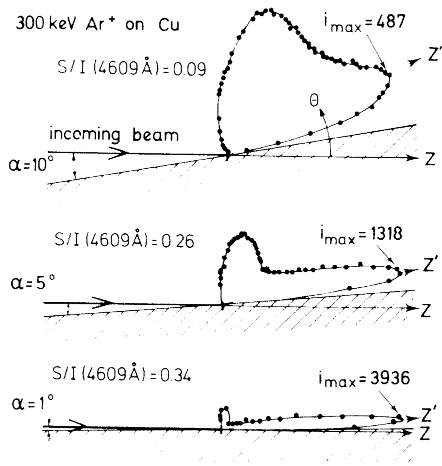

FIG. 2. Polar diagrams of the scattered-ion current at three angles of inclination α. Relative values of the maximum scattered current are given ($i_{\rm max}$, in arbitrary units), and each curve is normalized such that $i_{\rm max} = 1$ for that curve. Inserted are S/I values measured in the 4609-Å light emission at the corresponding angles α.

results of the scattered ion current for Ar^+ on a Cu target are shown in Fig. 2. We find in all cases for $\alpha \leq 5°$ a well-defined forward-scattered peak at $\theta \sim 2\alpha$ which we attribute to predominantly singly charged scattered beam ions[8] because no Ar III lines could be observed. The further peak near $\theta = 70°$ is assigned to sputtered Cu ions since it increases with incident ion mass, in agreement with sputtering yields,[6] and since weak Cu II lines are observed. In Fig. 2 measurements of S/I for the 4609-Å line are also inserted. From the rapid decrease of S/I with increasing θ we conclude that the interaction leading to the large orientations is a strong funtion of θ.

In a second stage in understanding of the anisotropic excitation process, one can draw conclusions from the magnitudes and signs of the measured S/I values. Assming LS coupling for the beam and target atoms the spin-orbit interaction is essentially turned off during the short interaction time ($<10^{-13}$ sec) of the ions with the surface. Thus, an oriented total orbital angular momentum $\langle\vec{L}\rangle$ can be generated by pure ion-surface Coulomb interaction processes. When the ion is leaving the surface, the corresponding isotropic spin S has to be coupled to the oriented $\langle\vec{L}\rangle$ in order to form all allowed fine-structure eigenstates $(LS)J$ which then can decay radiatively within a multiplet to lower states $(L_0S)J_0$. Under such circumstances S/I is only determined by the initial irreducible spherical "orbital" density matrix components[9,10] $\rho_q^{(k)}$, which are constrained by reflection symmetry with respect to the yz plane: $\rho_q^{(k)} = (-1)^k \rho_{-q}^{(k)}$.[11] This leads to

$$S/I = -\rho_1^{(1)} \begin{Bmatrix} JJ1 \\ 11J_0 \end{Bmatrix} \begin{Bmatrix} JJ1 \\ LLS \end{Bmatrix} \left(\rho_0^{(0)} \left(\frac{1}{3}\right)^{1/2} \begin{Bmatrix} JJ0 \\ 11J_0 \end{Bmatrix} \begin{Bmatrix} JJ0 \\ LLS \end{Bmatrix} + \left[\frac{1}{2}\rho_2^{(2)} - \left(\frac{1}{24}\right)^{1/2} \rho_0^{(2)}\right] \begin{Bmatrix} JJ2 \\ 11J_0 \end{Bmatrix} \begin{Bmatrix} JJ2 \\ LLS \end{Bmatrix} \right)^{-1}. \tag{1}$$

The four unknown $\rho_q^{(k)}$ components in this expression have been adjusted such that the measured $(S/I)_{\rm exp}$ are best reproduced by $(S/I)_{\rm theor}$, calculated with a single set ($\rho_0^{(0)} = 1$; $\rho_1^{(1)} = -0.5$; $\rho_0^{(2)} = 0.2$; $\rho_2^{(2)} = 0.15$) for all lines in Table I. The good agreement allows us to deduce from $\rho_1^{(1)} = -0.5$ that a unique orbital angular momentum $\langle -L_x \rangle$ is generated for all excited states in Table I.

The large orientation obtained with this effect suggests its use for level-crossing and magnetic-resonance studies of excited ionic levels. In order to demonstrate such an application we show in Fig. 3 the zero-field level-crossing (Hanle) signals obtained for the $4p'\,^2F_{7/2}$ state in the 4609-Å emission. Following the above discussion, a treatment similar to Ref. 10 leads to the expressions in Fig. 3 for the intensities at $B = 0$ and $B \to \infty$. Together with the deduced set of $\rho_q^{(k)}$ components, these expressions account sufficiently well for the measured intensities and explain the asymmetry with respect to the broken center line by the $\rho_2^{(2)}$ contribution from a finite alignment[9] of the excited state. In principle, the width of such Hanle signals can also yield a value for the lifetime τ of the excited state. However, the finite observation time due to the unknown scattered-ion velocity and the observation window leads to uncontrollable broadening (factor ~2 in Fig. 3) of the unperturbed line shape, so that in the present geometry lifetime determinations are not possible. But since this broadening does not affect the position of high-field level crossings (HFLC), this new technique can easily be extended to HFLC measurements of fine or hyperfine structures of ions and offers there a number of advantages owing to the indestructible nature of the targets, the large orientation, and the large magnetic fields which can be used along z' with minimal beam bending.

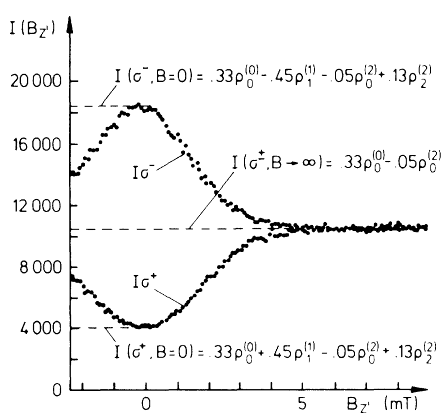

FIG. 3. Zero-field level-crossing signals for $I\sigma^-$, right circularly polarized light, and $I\sigma^+$, left circularly polarized light. For the formulas, see text.

In conclusion we could show that the ions leave the surface predominantly in the forward direction for small angles of incidence α, and that aside from surface cleanness such small scattering angles θ leads to large orientations in many excited states. Asuming LS coupling, we could interpret all S/I values by the generation of a uniquely oriented total orbital angular momentum, $-L_x$, of the excited states by Coulomb interaction with the surface. For a fuller understanding of the excitation process, however, further experiments with other ions, other excited-state configurations, and targets with better-controlled surface qualities are required. Such systematic studies, including scattering angular and energy dependences of S/I, could then also be applied to the analysis of solid-state properties of the targets, an example being the pickup of spin-polarized electrons[12] from magnetized media. As a further application we expect this excitation scheme to serve as a new tool in HFLC studies in atomic physics.

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Classical Theory of a Free-Electron Laser*

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We present a completely classical analysis of the small-signal regime of a free-electron laser. It is explicitly shown that the amplification is due to stimulated scattering produced by a bunching of the electron distribution.

Stimulated emission of bremsstrahlung in a transverse periodic magnetic field[1] has been recently observed by Elias *et al.*[2] In this experiment, a relativistic electron beam ($E=\gamma mc^2\simeq 24$ MeV) was passed through a tube centered within a superconducting double helix supplying a periodic static field. Amplification was achieved for the stimulated radiation in the direction of the electron beam. The stimulated emission of radiation in a transverse magnetic field has been analyzed by Dreicer,[3] Pantell, Soncini, and Puthoff,[4] Madey and co-workers,[5,6] and Sukhatme and Wolff.[7] All of these theories are quantum mechanical in nature. They give the impression that they have to be so, since it is argued that it is the electron recoil $\Delta p=h/\lambda_c$, where λ_c is the Compton wavelength, which is the source of a finite gain. Furthermore, quantum approaches, while agreeing on the structure of the gain formula, differ from one another by orders of magnitude in numerical coefficients.

In this Letter, we show that this problem is completely classical, and that the gain is produced by a bunching of the electron density in the presence of a field.

In order to avoid difficulties in numerical coefficients, we choose to work directly in the laboratory frame, and stay in a space-time description of the problem. We use the Weizsäcker-Williams approximation,[8] which allows us in the extreme relativistic limit to replace the static magnetic field of period λ_q by a pure electromagnetic field of wavelength

$$\lambda_i=(1+\beta)\lambda_q\simeq 2\lambda_q, \tag{1}$$

where

$$\beta^2\equiv 1-1/\gamma^2. \tag{2}$$

We describe the motion of the electron distribution $f(\vec{x},\vec{P},t)$ by the collisionless relativistic Boltzmann equation[9]

$$\frac{df}{dt}=\frac{\partial f}{\partial t}+\dot{x}_i\frac{\partial f}{\partial x_i}+\dot{P}_i\frac{\partial f}{\partial P_i}=0, \tag{3}$$

where $\vec{P}$ is the canonical momentum, $\vec{x}$ the position, and a dot expresses the total derivative with respect to time. The total number of electrons $N(t)$ is

$$N(t)=\int d^3x\int d^3P f(\vec{x},\vec{P},t). \tag{4}$$

The Boltzmann equation is coupled via the transverse current $\vec{J}_T$ to the Maxwell equations

$$+\nabla^2\vec{A}-c^{-2}\partial^2\vec{A}/\partial t^2=-\mu_0\vec{J}_T, \tag{5}$$

where $\vec{A}$ is the vector potential, and

$$\vec{J}_T(\vec{x},t)\equiv e\int d^3P\,\vec{v}_T f(\vec{x},\vec{P},t). \tag{6}$$

Here, e is the electron charge, and $\vec{v}_T$ the transverse component of the electron velocity.

In order to simplify the set of Eqs. (3) and (5), we now consider a model in which the electromagnetic field is transverse and depends on z and