the $J=\frac{3}{2}$ to $J=\frac{5}{2}$ transition in hydrogen, $n=4$. The sensitivity of quantum beats to the coherent population in a particular excited state provides possibilities for many interesting experiments. We are presently attempting to observe the two-photon[9] $P_{1/2}$-$P_{3/2}$ transition. This transition would be difficult to observe using normal rf spectroscopy because of the expected nearly equal initial populations and identical decay paths of the $P_{1/2}$ and $P_{3/2}$ levels. The quantum beats from the $P_{1/2}$-$P_{3/2}$ coherence are, however, expected to display resonant features similar to those described in this paper. Additionally, line narrowing may be achieved (without the necessity of two-phased rf regions[10]) by applying a single rf field to the coherent population for a time longer than a natural lifetime.

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Polarization of X Rays from Laser-Produced Plasmas*

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The first measurement of polarized x-rays from a laser-produced plasma is reported. The degree of polarization is related to the net anisotropy of the hot-electron velocity distribution.

Recent experiments[1] and theoretical calculations[2] have shown that the velocity distribution of hot electrons in laser-produced plasmas is probably anisotropic. In such cases, the free-free electron-ion bremsstrahlung should be polarized.

The first experimental measurement of the polarization of free-free bremsstrahlung in a plasma with an anisotropic velocity distribution was reported by Greene, Shohet, and Raimbault[3] in the case of a steady-state mirror-confined plasma, produced by electron cyclotron heating. A subsequent attempt to make a similar measurement in the case of laser-produced plasmas yielded inconclusive results.[4]

It is the purpose of this Letter to report the first successful measurement of the polarization of the x radiation from laser-produced plasmas, and to show the relationship between the anisotropy of the velocity distribution and the degree of polarization of the x rays.

The experimental apparatus is shown in Fig. 1. A 5-J Nd-glass laser was used to produce plasma using a flat polyethylene target. The laser system consists of a mode-locked Nd:YAlG (yttrium aluminum garnet) oscillator (5 mm in diameter

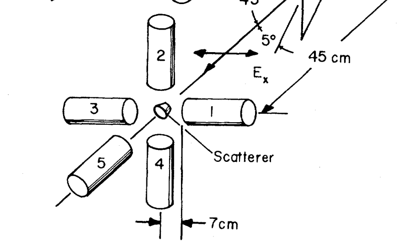

FIG. 1. Experimental arrangement of laser, target, and polarimeter.

and 75 mm in length) from which one 30-psec pulse is switched out and amplified by two Nd :YAlG rods (9.5 mm × 83 mm) followed by 19 mm × 500 mm and then 51 mm × 500 mm Nd:glass amplifier rods. The $f/3.5$ focusing optics yielded incident intensities of the order of 10^{15} W/cm^2 at the target. An 8% energy prepulse was introduced 30 psec or more in advance of the main laser pulse in all the target experiments.

The polarimeter shown in Fig. 1 is essentially that used in Ref. 4 except that the low-Z polyethylene scatterer has been made somewhat thicker, to provide a higher scattering efficiency for x-ray photons. The device incorporates five scintillator-photomultiplier detectors consisting of 1-mm NaI crystals with 0.05-mm Be windows mated to RCA 4516 PM tubes. Detector No. 5 is arranged to detect unscattered x rays through a small hole in the scatterer. The remaining detectors are divided into two pairs, forming two separate polarimeters; one pair has covering foils of 0.1-mm Al. X rays incident on the scatterer which are polarized horizontally are scattered vertically, due to the Compton effect, into the top and bottom detectors (No. 2 and No.4) while vertically polarized x rays are scattered horizontally into the two side detectors (No. 1 and No. 3). The Al absorbers result in a lower energy cutoff for the x rays of about 6 keV. An upper energy cutoff for detection of the x rays is at about 200 keV where photoabsorption in the 1-mm NaI crystals drops to 10%.

In order to select an observation angle at which significant polarization might be measured, some initial assumptions regarding the nature of the hot-electron velocity distribution must be made. These were that the distribution was either an anisotropic Maxwellian with the "temperature" parallel to the target normal greater than the "temperature" perpendicular to the target normal, or a drifting Maxwellian with drift velocity parallel to the target normal superimposed on an isotropic Maxwellian distribution function. Free-free electron-ion bremsstrahlung would produce x rays of varying intensity and polarization depending upon the angle of observation relative to the target normal. The intensity, and more strongly, the polarization are affected not only by the anisotropy of the electron velocity distribution, but also according to whether the x rays are produced by small- or large-angle scatterings, and whether the electrons are relativistic or nonrelativistic. It is believed that in this experiment the free-free transitions are large-angle scatterings experienced by nonrelativistic electrons. Accordingly, given the distribution functions mentioned, the x-ray polarization should be predominantly parallel to the target normal.

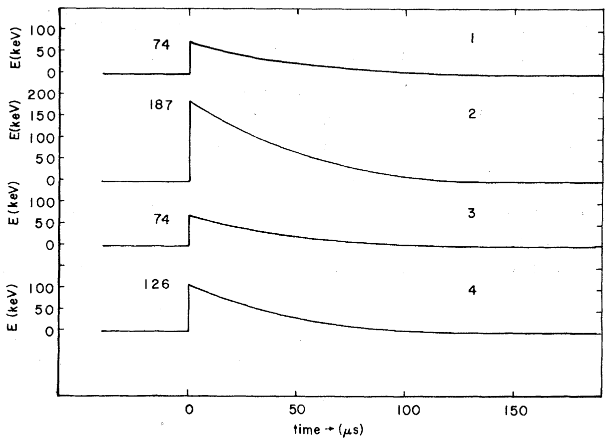

FIG. 2. X-ray intensity measured by the four detectors for a target shot at a 5° observation angle. Heights indicated in keV at $t = 0$ denote total x-ray energy recorded. Signals decay exponentially with the integrator time constant of 50 μsec.

Figure 2 shows experimental results for a shot in which polarization was measured at an observation angle 5° with respect to the target surface. The traces correspond to signals measured by calibrated detectors Nos. 1–4. Detectors Nos. 2 and 3 have the Al absorbers. Note that the signals on detectors Nos. 2 and 4 are larger than those on detectors Nos. 1 and 3, respectively. This implies that the x rays are polarized predominantly parallel to the target normal. A comparison between the unscattered and scattered x-ray intensities indicated a scattering efficiency of roughly 1%. Since the x rays all arrive at basically the same time, a rough estimate of the number of photons arriving at the detectors can be made, provided that the detector sensitivities are known. Single photons could be detected, but in the case shown here, all the detectors were measuring at least some tens of photons. The degree of polarization seems to be roughly the same for either the Al-covered or the uncovered detectors.

The intensity of x radiation produced per shot depends on the energy of the laser pulse. In fact, x-ray intensity appears to be greatest for laser energies below the highest energy available from the system. This may be due to self-focusing of the laser beam at the higher energies, thus cutting down the intensity of laser light striking the target. Given a sufficiently large measured x-ray intensity, the polarization was always parallel for the orientation shown in Fig. 1.

Two additional angles of observation were used. The first was such that the polarimeter observed x rays emitted along the target normal. In this configuration, no trend in the polarization was observed. The second orientation placed the polarimeter so that it observed x rays emitted parallel to the target surface but perpendicular to the plane of the laser beam and the target normal. The indicated polarization again appeared to be parallel to the target normal.

To determine the relation between velocity-distribution anisotropy and x-ray intensity and polarization, a Monte Carlo integration was performed to obtain x-ray fluxes for two orthogonal polarizations using cross sections for free-free electron-ion bremsstrahlung. Figure 3 shows the coordinate system and defines quantities used in the calculation. The differential scattering cross section $d\sigma$ for free-free electron-ion bremsstrahlung of nonrelativistic electrons, taken from the results of Bethe and Salpeter[5] is

$$d\sigma = A\frac{p}{p_0}\frac{1}{h\nu}\frac{(\vec{p}_0\cdot\hat{j}-\vec{p}\cdot\hat{j})^2}{|\vec{p}_0-\vec{p}|^4}\sin\theta\,d\theta\,d\gamma, \tag{1}$$

where A is a scale factor. For a fixed angle of observation φ, the integration is performed over the angles α, β, γ, and θ. These correspond to the angles of the incident- and scattered-electron momenta. The degree of polarization P is defined by $P=(J_1-J_2)/(J_1+J_2)$, where J_1 and J_2 are the fluxes of x-ray photons in the directions $\hat{j}_1$ and $\hat{j}_2$, respectively. $\hat{j}_1$ and $\hat{j}_2$ are orthogonal unit vectors. The fluxes J_1 and J_2 are obtained by use of the following formula

$$J_{1,2}=\int_{\theta,\gamma}d\sigma_{1,2}\int_{p_{\min}}^{\infty}\int_0^{\pi}\sin\alpha\,d\alpha\int_0^{2\pi}d\beta\,p_0^3 f(p_0,\alpha), \tag{2}$$

where $p_{\min}=(2mh\nu)^{1/2}$ and $d\sigma_1$ and $d\sigma_2$ are found by evaluating Eq. (1) with $\hat{j}$ set equal to $\hat{j}_1$ or $\hat{j}_2$, respectively. The intensity $[I=\pi(J_1+J_2)]$ and the degree of polarization are plotted as functions of x-ray photon energy in Fig. 4 for several angles of observation using the drifting Maxwellian and anisotropic Maxwellian distribution functions. The polarization is seen to approach zero while the intensity decreases slightly as the angle of observation approaches the target normal. Also, the polarization reverses sign when the photon energies become small compared to the electron thermal energies. No significant difference in the results is seen between the two types of distribution functions.

By computing both average polarization from the experiment and a comparable average from the theory for a range of perpendicular temperatures and anisotropies (anisotropic Maxwellian distribution function assumed) at the 5° observation angle, we conclude that the experimental polarization data, to be consistent with earlier Nd:glass laser experiments[6] ($T_{\text{hot}}\approx$ 8–10 keV), im-

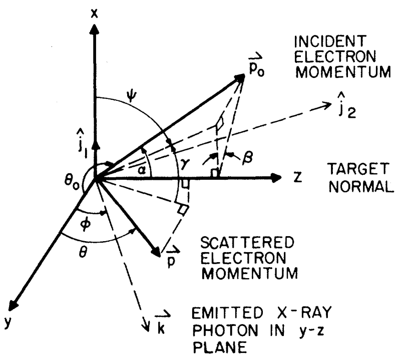

FIG. 3. Geometry and definitions for theoretical calculation.

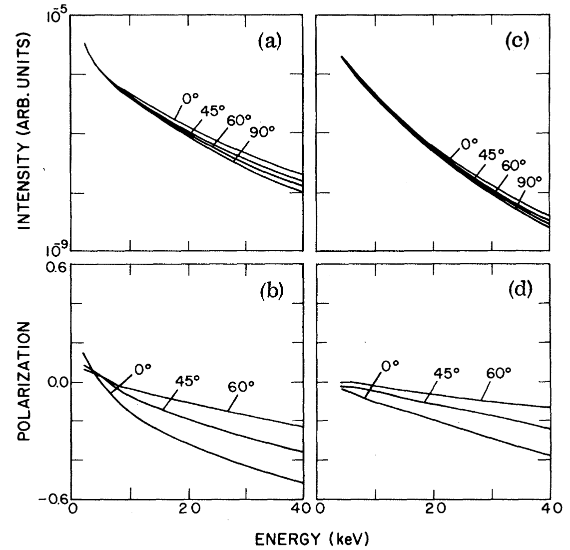

FIG. 4. Theoretical x-ray intensity and polarization vs energy for several observations angles. Distribution functions used are (a), (b) a drifting Maxwellian: $f(p_0,\alpha)=\exp\{-p_0^2/2mkT[\sin^2\alpha+(\cos\alpha-p_{\text{av}}/p_0)^2]\}$ with $p_{\text{av}}=(8mkT)^{1/2}$ and $kT=10$ keV; (c), (d) an anisotropic Maxwellian: $f(p_0,\alpha)=\exp\{-p_0^2/2mkT_\perp[\sin^2\alpha+T_\perp/T_\parallel)\cos^2\alpha]\}$ with $T_\perp/T_\parallel=0.5$ and $kT_\perp=10$ keV.

plies a net anisotropy $T_{\perp}/T_{\parallel} \approx 0.2$–$0.3$. Such anisotropy is in reasonable agreement with particle simulation results.[2,7] and may indicate that resonant absorption[7] is the process by which laser light is absorbed and hot electrons produced.

In summary, the polarization of the emitted x rays from laser-produced plasmas has now been measured successfully. The degree of polarization is related to the nature of the electron velocity distribution and is a much stronger function of distribution-function anisotropy than is the x-ray intensity. In addition, the polarization has been shown to be a quantity measurable at a *single angle* of observation while the intensity must be measured at at least *two separate* angles.

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Variation of Surface Reaction Probability with Reactant Angle of Incidence: A Molecular Beam Study of the Asymmetry of Stepped Platinum Crystal Surfaces for H-H Bond Breaking

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A molecular beam of H_2 and D_2 was used to investigate the angular dependence of the reaction probability for H_2-D_2 exchange ($H_2 + D_2 \rightarrow 2HD$) on the Pt(111) and Pt(*S*)-[6(111) × (111)] surfaces. On the stepped surface, a marked increase in the production of HD is observed when the beam of reactants strikes the open side of the step structure. The Pt(111) surface exhibits a smooth decrease in HD production from normal incidence to grazing angles.

In this Letter, we demonstrate for the first time how the reaction rate ($H_2 + D_2 \rightarrow 2HD$) depends on the angle of approach of the reactants to the atomic step structure on a high-Miller-index surface, Pt(332). The exchange reaction has been studied as a function of incident azimuthal angle (φ) and angle of incidence (θ). These results are compared with those obtained on a Pt(111) surface under identical conditions.

The importance of structural defects such as steps and kinks for catalytic bond breaking on single-crystal surfaces has been emphasized in recent years.[1] Since these sites are asymmetrical,[2,3] the reaction probability (efficiency of adsorption, bond breaking, etc.) may be determined by the direction of approach of the reactant molecule. The dependence of the reactivity at a surface site on the angle of approach of the reactant can only be investigated with a directed flow of molecules to the surface. The ultrahigh-vacuum molecular-beam-surface-scattering apparatus used in this study has been previously described in detail.[4] A mixed molecular beam of H_2 and D_2 is chopped before impinging on the platinum single-crystal surface. Periodic pulses of both the reaction product (HD) and the unreacted species (H_2 and D_2) are emitted from the surface and detected by a rotatable quadrupole mass spectrometer.

The detection was performed using two meth-