

FIG. 2. Narrow two-photon Doppler-free resonances of  $v = 0$  to  $v = 2$  in NO.

the 150-m% incident CO laser radiation the signal corresponded to an absorption of about 1%.

The technique of using a wave guide to do twophoton spectroscopy provides a means of obtaining high-field intensities over long path lengths with lasers of relatively weak powers. It should be mentioned that for the oversized wave guide  $(d/\lambda = 100)$  used here, the phase velocity does not significantly depart from its value in free space. Furthermore, with propagation in the wave guide, Doppler-free resonances can be observed over a

sizable absorption path without a residual Doppler broadening due to misalignment of the two oppositely propagating beams or angular beam spread arising from diffraction (which can occur in free-space propagation).

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 $^{1}$ M. A. Guerra, S. R. J. Brueck, and A. Mooradian, IEEE J. Quant. Electron. 9, <sup>1157</sup> (1973).

 $2^2$ M. D. Olman, M. D. McNelis, and C. D. Haure, J. Mol. Spectrosc. 14, 62 (1964).

 ${}^{3}$ T. C. James and R. J. Thibault, J. Chem. Phys. 41. 2806 (1964).

 ${}^{4}$ J. J. Gallagher and C. M. Johnson, Phys. Rev. 103, 1727 (1956).

<sup>5</sup>P. G. Favero, A. M. Mirri, and W. Gordy, Phys. Rev. 114, 1594 (1959).

 ${}^{6}$ J. H. Van Vleck, Rev. Mod. Phys. 23, 213 (1951). An  $h\nu = 69886$  cm<sup>-1</sup> is used to derive the theoretical values listed here; this value is chosen in order to be consistent with Gallagher and Johnson's values for the ground-state A-doublimg constants.

## Polarization of Target  $K$  X-Rays\*

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We point out that the  $K\alpha$  satellite spectra of Al target x rays exhibit strong polariza-We point out that the  $K\alpha$  satellite spectra of Al target x rays exhibit strong polariza zation in ion-atom collisions. The  $K\alpha'$  ( ${}^1P_1$ <sup>+1</sup>S<sub>0</sub>),  $K\alpha_3$  ( ${}^3P_{2,1,0}$ <sup>+3</sup> $P_{2,1,0}$ ), and the  $K\alpha_4$ (attion in ion-atom collisions. The  $\Lambda \alpha$  ( $\gamma_1 \gamma_2$ ,  $\lambda_0$ ),  $\Lambda \alpha_3$  ( $\gamma_2$ <sub>1,1,0</sub>),  $\gamma_1$ , and the  $\Lambda$ <br>( $\gamma_1$  +  $D_2$ ) transitions are, respectively, (+52 + 5)%, (-9+ 5)%, and (+9+ 5)% linearly polarized following 1.9-MeV He<sup>+</sup> bombardment and  $(+28\pm5)\%$ ,  $(-6\pm5)\%$ , and  $(+6\pm5)\%$ linearly polarized following  $1.9-MeV$  H<sup>+</sup> bombardment.

The polarization of characteristic x radiation by proton and electron impact has been discussed frequently in the last several years.<sup>1-8</sup> Mehlhorn<sup>3</sup> has suggested that electron and proton impact should cause alignment of the target atoms with respect to the beam  $(z)$  axis, in which case the resulting x radiation should have a nonisotropic angular distribution and should be polarized  $(n,$  $l > 0$ ,  $j > \frac{1}{2}$ . It was suggested that an important test of alignment would be the study of the polarization of  $L\alpha_1$  (3 $d_{5/2}$ -2 $p_{3/2}$ ) and  $L\alpha_2$  (3 $d_{3/2}$ -2 $p_{3/2}$ ) radiation. Hrdy, Hennis, and Bearden' did in fact observe a polarization of 14% for  $L\alpha_1$  in Hg. The characteristic  $K$  x radiation is in general not polarized since it is due to the decay of  $a j = \frac{1}{2} v a$ cancy state. However an important test case, which to the best of our knowledge has been com-

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pletely neglected, is the polarization of the first  $single-K$ -, multiple-L-shell-vacancy satellite. This satellite is due to the decay of the  $(1s)^{-1}(2p)^{-1}$ double-vacancy configuration' which couples to two terms  ${}^{1}P_1$  and  ${}^{3}P_{2,1,0}$ . These terms decay via three multiplets to the  $(2p)^{-2}$  final vacancy configuration and are labeled as follows:  $K\alpha'$  ( ${}^1P_1 \rightarrow {}^1S_0$ ),  $K\alpha_3$  ( ${}^3P_{2,1,0}$  +  ${}^3P_{2,1,0}$ ), and  $K\alpha_4$  ( ${}^1P_1$  +  ${}^1D_2$ ). An ideal system for the study of the polarization of these multiplets is a  $He$  beam on an Al target where the intensity of the first satellite is of the same order of magnitude as the intensity of the  $K\alpha_{1,2}$  line.

In this Letter we report the first observation of polarization of target  $K$  x radiation from an ionatom collision. Polarization measurements of this type should be an important test of collision



FIG. 1. Al  $K\alpha$  satellite spectra for 1.9-MeV He<sup>+</sup> bombardment. Upper spectrum: Rowland circle coplanar with beam axis,  $I_{\perp}$ . Lower spectrum: Rowland circle normal to beam axis,  $I_{\parallel}$ . These spectra yield polarizations of  $(52 \pm 5)\%$  for the  $K\alpha'$  transition,  $(-9)$  $\pm$  5)% for the  $K\alpha_3$  transition, and (+9 $\pm$  5)% for the  $K\alpha_4$ transition.

theories since the populations of the different substates must be correctly predicted to account for<br>the alignment of target atoms as manifested in<br>the polarization of the emitted radiation.<sup>1,2</sup> the alignment of target atoms as manifested in the polarization of the emitted radiation. $1.2$ 

The polarization fraction is defined  $as^{1,2}$ 

$$
P = (\mathcal{G}_{\parallel} - \mathcal{G}_{\perp}) / (\mathcal{G}_{\parallel} + \mathcal{G}_{\perp}), \qquad (1)
$$

where  $\mathcal{G}_{\parallel}(\mathcal{G}_{\perp})$  is the intensity of the radiation propagating in the  $x$  direction with the electric vector parallel (perpendicular) to the beam,  $z$ , direction. With the use of a 4-in. curved crystal

spectrometer as a polarimeter, the measured intensities  $I_{\parallel}$  (Rowland circle normal to the beam axis) and  $I_{\perp}$  (Rowland circle coplanar with the beam axis) must be corrected to give the polarization fraction. Under the assumption of an imperfect or mosaic crystal the polarization fraction is given in terms of the measured intensities  $as^{10}$ 

$$
P = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + I_{\perp}} \frac{1 + \cos^2(2\theta)}{1 - \cos^2(2\theta)},
$$
\n(2)

where  $\theta$  is the Bragg angle. We take the values  $I_{\parallel}(I_{\perp})$  to be the intensities of a given multiplet relative to the intensity of  $K\alpha_{1,2}$  which is assume unpolarized.

Figure 1 displays the Al spectra observed in the two orientations of the spectrometer for 1.9- MeV He on Al. The spectra were taken with a Johansson-type ammonium dihydrogen- phosphate (ADP) curved crystal. The entrance aperture to the spectrometer was a cylindrical-tubular collimator. The characteristic  $K\alpha_{1,2}$  line, as well as the first two single- $K-$ , multiple- $L$ -ionization satellite structures, is included in the energy region shown. The spectra are plotted in units of the integrated intensity of the  $K\alpha_{1,2}$  line divide by 100. The most obvious spectral difference is the K $\alpha'$  relative yield. The intensity in the  $I_{\parallel}$  orientation is greatly enhanced compared to the intensity in the  $I_{\perp}$  orientation.

The spectra were fitted with a least-squares fitting program to obtain the relative intensities of the peaks  $K\alpha'$ ,  $K\alpha_3$ , and  $K\alpha_4$  relative to  $K\alpha_{1,2}$ . The results are tabulated in Table I for both He' and  $H^+$  bombardment of Al. The measured polarizations of  $K\alpha'$ ,  $K\alpha_3$ , and  $K\alpha_4$  are +52, -9, and  $+9\%$  for He<sup>+</sup> bombardment and  $+28$ ,  $-6$ , and  $+6\%$ , respectively, for  $H^+$  bombardment. The Bragg angles for the transitions occur at 102.62, 102.31, and 102.03°, respectively, for an ADP crystal.

It is interesting to compare the observed relative line strengths of the multiplets of the first

Projectile			Ratio to $K\alpha_{1,2}$ (%)		$I_{\parallel}-I_{\perp}$ $I_0 + I_1$	$1 + \cos^2(2\theta)$	₽
(Energy)	Line	Initial $\rightarrow$ final	Ι,	$I_{\rm H}$	(%)	$1-\cos^2(2\theta)$	(%)
$He^+$ (1.9 MeV)	$K\alpha'$	${}^{1}P_1 \rightarrow {}^{1}S_0$	6.5	18.2	$+47.5$	1.100	$+52 \pm 5$
	$K\alpha_{3}$	$\mathbf{A} \rightarrow 3P_{2,1,0}$ <sup>3</sup> $P_{2}$ <sub>1</sub> , 0 <sub>1</sub> + 1 <sub>1</sub> $D_2$	143.6	122.1	$-8.1$	1.095	$-9 \pm 5$
	$K\alpha_A$		43.1	51.2	$+8.6$	1.091	$+9+5$
$H^+$ (1.9 MeV)	$K\alpha'$	${}^{1}P_{1} \rightarrow {}^{1}S_{0}$	0.66	1.11	$+25.4$	1.100	$+28 \pm 5$
	$K\alpha_{3}$	${}^3\!P_{\,2\,,1\,,0}$ $\cdot\,{}^3\!P_{\,2\,,\,1\,,\,0}$	11.66	10.50	$-5.2$	1,095	$-6 \pm 5$
	$K\alpha_A$	${}^{1}P_1 \rightarrow {}^{1}D_2$	3.97	4.44	$+ 5.6$	1.091	$+ 6 = 5$

TABLE I. Polarization data.



satellites to their predicted line strengths assuming statistical populations. These results are given in Table II where the total yield of the three multiplets in the satellite is normalized to 15 in each case. Note also that the intensities in the parallel and perpendicular orientations as given in Table I summed over the three multiplets are identical within the experimental uncertainty  $[e.g., (\sum I_{\parallel} - \sum I_{\perp})/(\sum I_{\parallel} + \sum I_{\perp}) = -0.004$  for He and  $-0.007$  for H<sup>+</sup>].

The polarization fraction, P, of each of three<br>transitions  $K\alpha'$  ( ${}^{1}P_1 \rightarrow {}^{1}S_0$ ),  $K\alpha_3$  ( ${}^{3}P_{2,1,0} \rightarrow {}^{3}P_{2,1,0}$ ) , of each of three<br>  ${}^{3}P_{2,1,0}$  +  ${}^{3}P_{2,1,0}$ , and  $K\alpha$ <sub>4</sub> ( ${}^{1}P_1$  +  ${}^{1}D_2$ ) are given in terms of the partial ionization cross sections for the  $L = 1$  states with  $M_L = 0$ ,  $\sigma_0$ , and with  $|M_L| = 1$ ,  $\sigma_1$ , by Fano and Macek' and Percival and Seaton' as

$$
P(K\alpha') = (\sigma_0 - \sigma_1) / (\sigma_0 + \sigma_1), \qquad (3)
$$

$$
P(K\alpha_3) = -3D(\sigma_0 - \sigma_1)/[\sigma_1(8+D) + \sigma_0(4-D)], \quad (4)
$$

$$
P(K\alpha_4) = (\sigma_0 - \sigma_1) / (7\sigma_0 + 13\sigma_1). \tag{5}
$$

From Eqs. (3)-(5) we see that the polarization fractions of  $K\alpha'$  and  $K\alpha_4$  have the same sign, fractions of  $K\alpha'$  and  $K\alpha_4$  have the same sign, whereas  $K\alpha_3$  has the opposite sign. This is in agreement with the observed polarization fractions. In Eq. (4), D is the depolarization<sup>11</sup> due to the superposition of the three possible  $J$  states of the  ${}^{3}P$  term. The range of the depolarization is given by the two limits  $\Delta \omega_{JJ'} \tau \ll 1$  for which  $D = 1$ and  $\Delta\omega_{JJ'}\tau \gg 1$  for which  $D = \frac{5}{18}$ . In these two limits the polarization fractions are given by

$$
P(K\alpha_3) = -(\sigma_0 - \sigma_1)/(\sigma_0 + 3\sigma_1)
$$
 (6)

for  $D = 1$  ( $\Delta \omega_{JI'} \tau \ll 1$ ) and by

$$
P(K\alpha_3) = -15(\sigma_0 - \sigma_1)/(67\sigma_0 + 149\sigma_1)
$$
 (7)

for  $D=\frac{5}{18}$  ( $\Delta \omega_{JJ}$ ,  $\tau \gg 1$ ). All of the above equations demonstrate that when the ionization cross section is independent of the substates,  $M$ , then the polarization fractions are zero. The  $K\alpha'$  transition can attain a +100% polarization for  $\sigma_1 = 0$  and a – 100% polarization for  $\sigma_0 = 0$ . The  $K\alpha_A$  transition can attain a maximum positive polarization of only +14% for  $\sigma_1 = 0$  and a maximum negative polarization of only  $-7.7\%$  for  $\sigma_0 = 0$ . Therefore

the polarization of  $K\alpha'$  is a more sensitive measure of the ratio of the partial ionization cross sections,  $\sigma_0/\sigma_1$ .

Using Eq. (3) and our experimental value of  $P(K\alpha')$  for He<sup>+</sup> bombardment, we obtain the ratio  $\sigma_0/\sigma_1 = 3.2$ . This ratio can in turn be used in Eqs. (4)-(7) to predict the polarization of  $K\alpha_s$  and  $K\alpha_s$ . From Eq. (5) we obtain  $P(K\alpha_A) = +6.2\%$  which is in agreement with our experimental value of  $(+9\pm 5)\%$ . From Eqs. (6) and (7) we obtain  $P(K\alpha_s)$  $(1 + \sigma \pm 3)$ ). From Eqs. (b) and (f) we obtain  $P(\Lambda \alpha_3)$ <br>= - 35.2% (for  $\Delta \omega_{JJ'}$ ,  $\tau \ll 1$ ) and  $P(K\alpha_3)$  = - 9.0% (for  $\Delta\omega_{JJ'}\tau \gg 1$ ) compared with our experimental value of  $(-9 \pm 5)\%$ . The calculated polarization using  $\sigma_0/\sigma_1 = 3.2$  is thus in agreement with the experimentally measured polarization in the  $\Delta \omega_{JI'} \tau \gg 1$ depolarization limit.

The H'-induced spectra can be analyzed in a similar manner. Using our experimental value of  $P(K\alpha')$  we obtain the ratio  $\sigma_0/\sigma_1 = 1.8$ . From Eq. (5) we thus predict  $P(K\alpha_4) = 3.1\%$ , which is also in agreement with our measured value of  $(+6)$  $\pm 5\%$ . From Eqs. (6) and (7) we obtain  $P(K\alpha_3)$  $= -14.6\%$  (for  $\Delta \omega_{JJ'}$ ,  $\infty$ ) and  $P(K\alpha_3) = -4.4\%$ (for  $\Delta \omega_{II}$ ,  $\tau \gg 1$ ) compared with our experimental value of  $(-6 \pm 5)\%$ . As for the He<sup>+</sup> case, the calculated polarization for the H<sup>+</sup> case using  $\sigma_o/\sigma$ . =1.<sup>8</sup> is in agreement with the measured polarization in the  $\Delta \omega_{JJ}$ ,  $\tau \gg 1$  depolarization limit.

We next discuss a mechanism which may give rise to the observed polarization. In the impactparameter formulation of ionization cross secparameter formulation of ionization cross sec-<br>tions,<sup>12</sup> the single-1s, single-2p ionization can be represented by

$$
\sigma_{1s,2b} = \sum_{M_L} 2\pi \int_0^\infty P_{1s}(b) P_{2b,M_L}(b) b \, db \,, \tag{8}
$$

where  $P_{1s}(b)$  is the probability of K-shell ionization as a function of impact parameter,  $b$ , and  $P_{2p, M_{I}}(b)$  is the probability of ionization of the 2p subshell in the  $M_L$  substate (where  $M_L = 0,+1$ ) of the 2p subshell. The probabilities  $P(b)$  of ionization are usually taken to be cylindrically symmetric about the beam axis  $(z \text{ axis})$  as implied by Eq. (8). However because of the different spatial charge distributions for the different substates, the  $P_{2p, M_r}(b)$ 's are different functions of b for different values of  $M_L$ . In the present situation the range of  $b$  is selectively small since we require 1s-shell ionization simultaneously with  $2p$ -subshell ionization. The charge distribution of the  $M_L = 0$  substate of the 2p wave function is in fact concentrated along the  $z$  axis which corresponds to small impact parameters, whereas the  $|M_L|$  =1 substate charge distribution is distributed over larger impact parameters. This qualitatively de-

scribes the polarization results obtained in this experiment.

The calculated polarization should take into account the single- $K-$ , single- $L$ -ionization amplitude due to the "shake" effect<sup>13</sup> in addition to the amplitude due to Coulomb ionization. We expect that the "shake" up process will not lead to polarization. Therefore, the larger the relative contribution to ionization due to "shake" the smaller the observed polarization.

We note the following points concerning the polarization in this particular system: (1) Target atoms are not subject to the multiple-collision problems of beam polarization experiments. (2) Cascading from higher states is not a problem as it is in a few-electron system. (3) The beam divergence due to scattering is much less for ion impact than for  $e^-$ -induced x-ray spectra. (4) The total  $K\alpha L^1$  satellite is polarized by  $\leq 1\%$ , indicating that large polarization effects may be hidden by inappropriate averaging. (5) Our measurements predict that the Auger emission from the  ${}^{1}P_1$  state should exhibit anisotropy.

In conclusion, we have made the first observation of the polarization of  $K$  x radiation emitted from stationary states of targets when bombarded by 1.9-MeV  $H^+$  and He<sup>+</sup> beams. The measurements are shown to be internally consistent in the prediction of the substate populations. These measurements can be used as a direct test of collision theories for inner-shell ionization.

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 $1$ U. Fano and J. H. Macek, Rev. Mod. Phys. 45, 553 (1973).

<sup>2</sup>I. C. Percival and M. J. Seaton, Philos. Trans. Roy. Soc. London, Ser. <sup>A</sup> 251, 113 (1958).

 ${}^{3}\text{W}$ . Mehlhorn, Phys. Lett. 26A, 166 (1968).

 $^{4}$ J. Hrd $\circ$ , A. Hennis, and J. A. Bearden, Phys. Rev. A 2, 1708 (1970). Note that the polarization fraction differs by sign as compared to the usual definition as given in Refs. 1-3.

 $5J.$  Eichler and W. Fritch, in *Proceedings of the Sec*ond International Conference on Inner Shell Ionization Phenomena, Freiburg, 1976, edited by W. Mehlhorn R. Brenn (Univ. of Freiburg, Freiburg, Germany, 1976), p. 21.

 ${}^{6}E.$  G. Berezhko and N. M. Kabachnic, in Proceedings of the Second International Conference on Inner Shell Ionization Phenomena, Freiburg, 1976,' edited by W. Mehlhorn and R. Brenn (Univ. of Freiburg, Freiburg, Germany, 1976), p. 23.

<sup>7</sup>W. Schmitt, W. Sander, and W. Mehlhorn, in  $Pro$ ceedings of the Second International Conference on Inner Shell Ionization Phenomena, Freiburg, 1976, edited by W. Mehlhorn, and R. Brenn (Univ. of Freiburg, Freiburg, Germany, 1976), p. 23.

 ${}^{8}$ B. Cleff and W. Mehlhorn, J. Phys. B 7, 605 (1974).  $^{9}P$ . Richard, Atomic Inner Shell Processes, Vol. I. Ionization and Transition Probabilities, edited by

B. Crasemann (Academic, New York, 1975), p. 73.

<sup>10</sup>B. E. Warren, *X-Ray Diffraction* (Addison-Wesley, Reading, Mass., 1969), p. 334.

<sup>11</sup>J. Macek, private communication. The depolarization factor is

$$
D = \sum_{JJ'} \frac{(2J'+1)(2J+1)}{(2S+1)} \left\{ \frac{J'J2}{LLS} \right\}^2 \frac{1}{1 + \Delta \omega_{J'J}}.
$$

where  $\Delta \omega_{J'J}$  is the fine-structure splitting and  $\tau$  is the lifetime of each of the states.

 $^{12}$ J. Bang and J. M. Hansteen, Kgl. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 31, No. 13 (1959).

 $^{13}$ T. Aberg, Atomic Inner Shell Processes, Vol. I. Ionization and Transition Probabilities, edited by B. Crasemann (Academic, New York, 1975), p. 353.