Polarization of argon *K* x radiation following electron-impact ionization

R. K. Singh and R. Shanker*
Atomic Physics Laboratory, Physics Department, Banaras Hindu University, Varanasi–221 005, India
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The angular and impact-energy dependence of the characteristic *K* x radiation of argon produced from collisions of electrons with 10.0–24.0 keV energies has been investigated. The radiation is found to show a net linear polarization of up to $(17\pm6)\%$ in the investigated range of impact energy. This anisotropy is believed to arise from the strong polarization of $K\alpha$ satellite lines of argon produced by the *simultaneous* decay of a single *K*- and multiple *L*-shell vacancy states created in the collisions. Also, the behavior of the net polarization of *K* radiation as a function of impact energy in the range of 10.0 to 24.0 keV has been studied. Over the range of impact energies, the polarization is found to show a nearly constant value within the experimental uncertainty.

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I. INTRODUCTION

Collisions of energetic electrons with "free" atoms may give rise to the emission of both characteristic and noncharacteristic x radiation. The characteristic part of the x-ray spectrum is due to the decay of inner-shell vacancies created in the collisions, while the noncharacteristic radiation (bremsstrahlung) is caused due to the deceleration of incident electrons in the Coulomb field of the target atoms. This atomic field bremsstrahlung has a high-energy cutoff at a photon energy $k=T$, the incident energy of the electron [1].

The polarization of the characteristic x radiation induced by electrons and protons has been discussed frequently in the past [2–5]. It has been suggested by Mehlhorn [3] that electron and proton impact should cause alignment (polarization) of the radiation emitted from the target atoms with respect to the incident beam direction, say, *z* axis; in which case the emitted radiation should have a nonisotropic angular distribution and should be polarized for states having n, $l>0$ and $j>\frac{1}{2}$. Hrdy *et al.* [6] did in fact observe a polarization of 14% for the $L\alpha_1$ radiation in Hg. The characteristic *K* x radiation is, in general, not polarized because it is emitted due to the decay of a $j=\frac{1}{2}$ vacancy state. However, observation of the polarization of $K\alpha$ satellite x rays of a thick Al target having values as large as 52% by 1.9 MeV He^+ and 28% by 1.9 MeV H^+ bombardment was reported by Jamison and Richard [7]. This large polarization was suggested to occur due to a simultaneous excitation of one *K* and multiple vacancies in the *L* shell of Al. Künst and Mehlhorn [8] recently measured the alignment of Xe^+ ions after L_3 ionization by 5–6 keV electrons with Ar *K* reference radiation. In these measurements the authors took the Ar $K\alpha$ radiation to be "isotropic." Löw *et al.* [9] studied the energy dependence of the *K*-shell satellite intensity of low-*Z* elements of the solid targets through x-ray detection under impact of 10–20 keV electrons. They found that the ratio of *L*-shell multiple ionization to *K*-shell single ionization, KL^n/KL^0, is an impact-energy-dependent quantity. Albiez *et al.* [10] and Thoma *et al.* [11] measured the KL_{23} double ionization of neon using Auger electron spectroscopy by electron impact and determined the alignment A_{20} of $KL_{23}(^1P,^3P)$ states for a small range of impact energies (1.5–4.0 keV). They concluded that the alignment of ^1P and ^3P states of the KL_{23} vacancy configuration of neon is zero within the experimental uncertainty and that the average value of A_{20} for $^{1,3}P$ is -0.06 ± 0.05 for the highest impact energy of 4.0 keV.

The angular distributions of the characteristic photon emission may be nonisotropic and can be characterized by a single parameter *P*, the degree of polarization [12],

$$I(\theta)/I(90°)=1-P\cos^2\theta, \tag{1}$$

where θ stands for the angle of photon emission with respect to the incident electron beam direction. Also, the intensity distributions of the radiation can be written [10,11] as

$$I(\theta)=\frac{I_0}{4\pi}[1+A_{20}\alpha_2P_2(\cos\theta)], \tag{2}$$

where A_{20} is the alignment of the initial double-vacancy state, α_2 is the x-ray decay parameter, $P_2(\cos\theta)$ is the second Legendre polynomial, and I_0 is the total intensity of the x-ray line. In view of the above results of different workers, it was considered worthwhile to see whether there is any significant effect of higher impact energies of electrons on the magnitude of polarization *P* for KL_{23} vacancies created in a "gaseous" target, such as argon.

In this article, we report the observation of a net linear polarization $P=(17\pm6)\%$ of the argon *K* x radiation emitted from collisions of electrons having energies between 10 and 24 keV with *free* argon atoms. The polarization fractions are determined by measuring the angular distributions of *K* x radiation using a Si *p-i-n* x-ray detector. Such measurements are important from the viewpoint of testing collision theories [13], as the population of different magnetic substates must be correctly predicted to account for alignment of the target atoms and should manifest in the emitted radiation [2].

*Corresponding author. Email address: rshanker@banaras.ernet.in

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II. EXPERIMENTAL METHOD

The present measurements were carried out on our recently developed experimental setup dedicated for studies of multiple-ionization processes in keV electron-atom/molecule collisions [14–16]. A well-focused beam of electrons ($\sim$1–20 μA, diameter $\sim$3 mm) entered a collision chamber and impinged on a thermal gas target effusing from a multicapillary tube ($\Phi=5$ mm) at 90°. The tip of the multicapillary tube was placed at about 5 mm above the incident beam. A visual checking of the electron beam spot was done using a fluorescent screen placed at the collision center. The scattering chamber was differentially evacuated by a 240 l/s turbomolecular pump; the base pressure of the chamber was maintained at a pressure better than 1×10^{-6} Torr. A high-resolution, thermoelectrically cooled Si *p-i-n* photodiode x-ray detector equipped with a charge-sensitive preamplifier (Model XR-100T) and a 12.5 μm Be window was employed to record the x-ray spectra. The energy resolution (full width at half maximum) of the detector was found to be 180 eV at 5.9 keV. The operating pressure of the target gas was kept at 2.4×10^{-4} Torr, to ensure the single-collision condition. The angular distributions of the target x rays were measured with the help of a collimator with a circular aperture of 3 mm in front of the detector kept inside the vacuum chamber and placed relatively close (about 6.0 cm) to the collision center. The x-ray detection was made within a cone with $\Delta\alpha=5°$ yielding a solid angle of 0.016% of 4π. The detector could be placed at different angles with respect to the incident beam direction, e.g., at 30°, 60°, 90°, 120°, and at 135° with an angular uncertainty of about $\pm1°$. Care was taken to reduce the slit-edge scattering of the incident beam and to prevent thick-target bremsstrahlung radiation, if produced in the chamber, from reaching the detector. The signals generated by the detector were processed by standard NIM modules and were interfaced with a 200 MHz. Pentium-based 4 K multichannel analyzer (Oxford PCA-3). The data acquisition, management, stripping, and plotting were done on this computer. Normal data collection times at a given emission angle and impact energy were about 40 min. A few sets of repeated runs of data at different angles were made to check and ensure the reproducibility of the results. A typical spectrum of argon *K*-shell x radiation produced in collisions of 14.0 keV electrons with argon gas is shown in Fig. 1. The spectra have been smoothed with five-point averaging and were fitted with a Gaussian fitting program to obtain the relative intensities of the *K*-line peaks on top of a bremsstrahlung continuum background. The total uncertainty involved in the *K*-line intensity measurements stemmed basically from the uncertainties in the beam current, beam integration, angle uncertainty, and data collection statistics; their individual contributions were found to be, respectively, <1%, 2%, 2%, and 3%. Thus, the total uncertainty of the angular distribution data is a little over 4%.

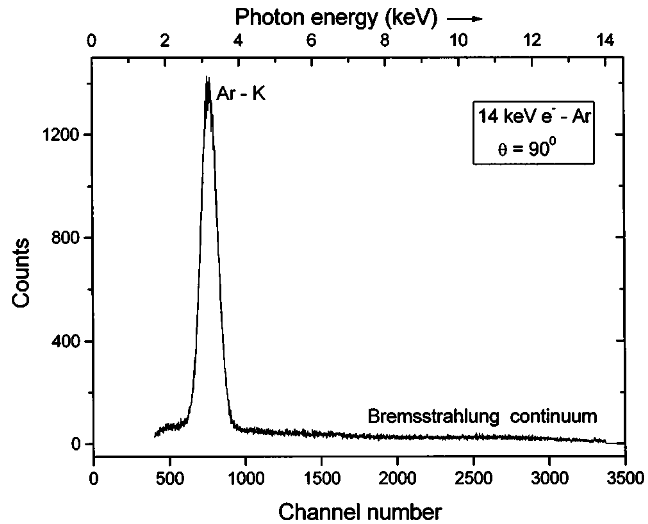

FIG. 1. A typical spectrum of Ar *K*-shell x radiation produced in collisions of 14.0 keV electrons with argon atoms. Angle of photon detection is 90° with respect to the incident electron beam direction.

III. POSSIBLE MECHANISMS FOR ARGON *K*-LINE POLARIZATION

Mechanisms responsible for the observed net linear polarization of *K* radiation of argon in the present investigation can be understood on the following lines.

The impact-parameter formalism of ionization cross sections for a simultaneous single *K* and single $2p$ ionization can be represented [17] by

$$\sigma_{1s,2p}=\sum_{M_L} 2\pi\int_0 P_{1s}(b)P_{2p,M_L}(b)b\,db \qquad (3)$$

where $P_{1s}(b)$ and $P_{2p,M_L}(b)$ are, respectively, the impact-parameter-dependent ionization probabilities of the K shell and L subshells ($2p$) with M_L magnetic substates having values equal to 0 and ±1. $P(b)$ is usually taken to be a cylindrically symmetric distribution about the beam axis (say, the z axis) as implied by Eq. (3). Moreover, the differential spatial charge distributions of different magnetic substates are different and the $P_{2p,M_L}(b)$ values are known to be different for different impact parameters b. The charge distribution of the $M_L=0$ substate of the $2p$ wave function will be concentrated along the z axis, while the charge distributions of the $M_L=\pm1$ substates will be over large values of impact parameters. This situation leads to an unequal population of $2p$ magnetic substates with a simultaneous population of the K shell ($1s^{-1}2p^6$) which is always isotropic. As a result, the radiation emitted due to KL_{23}-vacancy decay suffers a net polarization and consequently, the K-shell x-ray line shows a polarization (A_{20}) of a measurable magnitude.

The polarization of the first single-*K*-, multiple-*L*-shell vacancy satellite is expected to play a significant role in the net polarization of *K*-shell radiation. It may, however, be mentioned that our x-ray spectrometer is not capable of resolving these satellite lines in the *K* x-ray spectra. Moreover, the final effect of polarization of satellite lines should manifest in the net polarization of the *K* radiation. Let us consider the situation where the argon atoms in their ground state ($^1S_0$) are excited by energetic electrons such that one of the $2p$ electrons and one of the $1s$ electrons are simultaneously knocked out from their respective shells or subshells, leaving the par-

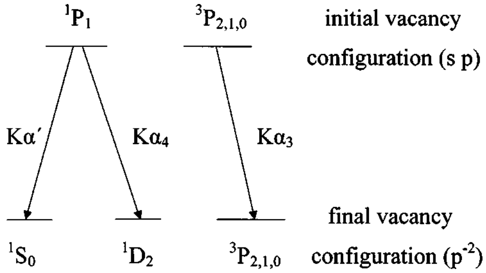

FIG. 2. X-ray transitions for $K\alpha$ satellite lines arising due to the simultaneous decay of a single K- and multiple L-shell vacancies in argon following electron-impact ionization.

ent atom in a doubly charged state $(1s^{-1}2p^{-1})$, i.e., Ar^{2+}. This doubly charged ion of the *sp* configuration will give rise to two terms: $^1P_1$ and $^3P_{2,1,0}$. These highly excited states of the ion can decay through emission of K x-ray photons to the terms $^1S_0$, $^1D_2$, and $^3P_{2,1,0}$ of a final vacancy configuration of $(2p)^{-2}$ type. The sequence of these reaction channels may be written as follows:

$$\text{(i)} \quad \mathrm{Ar}^0{:}1s^22s^22p^63s^23p^6\ {}^1S_0 \quad \text{(ground state)}, \quad (4)$$

$$\text{(ii)} \quad e^- + \mathrm{Ar}^0 \rightarrow \mathrm{Ar}^{2+}(1s^{-1}2p^{-1}) + 3e\uparrow \quad {}^1P,{}^3P \quad (sp\ \text{configuration}), \quad (5)$$

$$\text{(iii)} \quad \mathrm{Ar}^{2+}(1s^{-1}2p^{-1}) \rightarrow \mathrm{Ar}^{2+}(1s^22p^2) + \gamma \quad (K_\alpha\ \text{photons}) \quad {}^1P,{}^3P,{}^1D \quad (p^{-2}\ \text{configuration}). \quad (6)$$

The resulting x-ray satellite lines are

$$K\alpha'{:}\quad {}^1P_1 \rightarrow {}^1S_0 \quad \text{(with relative weight 1)}, \quad (7a)$$

$$K\alpha_3{:}\quad {}^3P_{2,1,0} \rightarrow {}^3P_{2,1,0} \quad \text{(with relative weight 9)}, \quad (7b)$$

$$K\alpha_4{:}\quad {}^1P_1 \rightarrow {}^1D_2 \quad \text{(with relative weight 5)}. \quad (7c)$$

The possible x-ray transitions between initial and final vacancy configurations induced by collision of energetic electrons with argon atoms are shown schematically in Fig. 2. These transitions result in three satellite lines, namely, $K\alpha'({}^1P_1 \rightarrow {}^1S_0)$, $K\alpha_3({}^3P_{2,1,0} \rightarrow {}^3P_{2,1,0})$, and $K\alpha_4({}^1P_1 \rightarrow {}^1D_2)$. They are obtained if recorded by a high-resolution spectrometer, for example, by a curved crystal spectrometer. In these spectra, it is generally expected that $K\alpha'({}^1P_1 \rightarrow {}^1S_0)$ is strongly polarized, while $K\alpha_3$ and $K\alpha_4$ are weakly polarized; the $K\alpha_{1,2}$ lines $(KL_1L_{2,3})$, the main diagram lines, are assumed to be unpolarized (see Ref. [7]). Polarization of $K\alpha'$ is a more sensitive measure of the ratio of the partial ionization cross sections σ_0/σ_1, where σ_0 and σ_1 are the partial ionization cross sections for $M_L=0$ and $|M_L|=1$, respectively.

Using the scaling law [18], one can obtain the individual charge state spectra [19,20] of ions produced as a result of

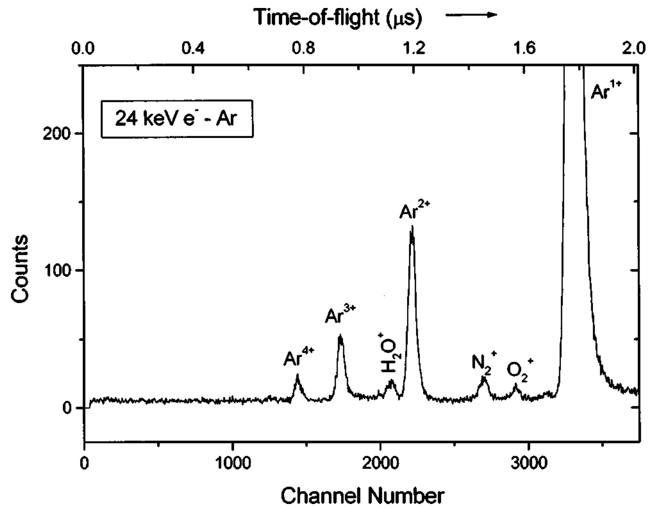

FIG. 3. Time-of-flight spectrum of argon ions resulting from collisions of 24.0 keV electrons with argon atoms. A time resolution of about 40 ns is seen for a 2 μs time window.

the ionization of each single atomic subshell and hence also partial cross sections for the production of ions in a charge state n in the νth subshell $(\nu = K, L, \dots)$. We have performed calculations for different contributions coming from ionization of various shells and subshells [21]. It is seen that a significant contribution to the final ionization comes from the K shell, L shell, and of course from the M shell in the production of ions of higher charge states. For example, in 10 keV e^--Ar collisions, the Ar^{4+} ions are produced due to 5.33% K-shell ionization, 70.35% L_1-subshell ionization, and 24.32% $L_{2,3}$-subshell ionization simultaneously. From the time-of-flight spectra that we have recorded very recently in our laboratory for studies of multiple ionization of argon atoms in collisions of 10.0 to 24.0 keV electrons with a dilute argon atomic target, it is observed that argon ions having charge states up to $+4$ are produced. These spectra are obtained by measuring coincidences between the ejected electrons (of all energies) at 90° to the incident beam direction and the multiply charged argon ions (see Fig. 3). The above calculations give excellent agreement between our experimental data [21] and different theoretical predictions for the relative charge state fractions as a function of the argon charge states. It may be pointed out here that the charge state spectra of argon ions can give information about the initial multiple vacancies if one considers the radiative and nonradiative decay modes quantitatively. It is well established that the fluorescence yields of K-shell-ionized argon is about 12% and the Auger yield is 88%. This means that the lone argon K vacancy is preferably filled through Auger transitions. Thus, for an initial one K and one $L_{2,3}$ vacancy, the final charge state of the ion will attain the minimum value of 3 by filling the K vacancy through an Auger transition. Thus, production of the final charge state of 3 (Ar^{3+}) will correspond in greater probabilities to an initial one K and one $L_{2,3}$ vacancy in argon. These initial vacancies (one K and one $L_{2,3}$) will also undergo radiative decay via emission of the $K\alpha$ x rays but with a smaller probability.

From the above consideration and from the spectra shown

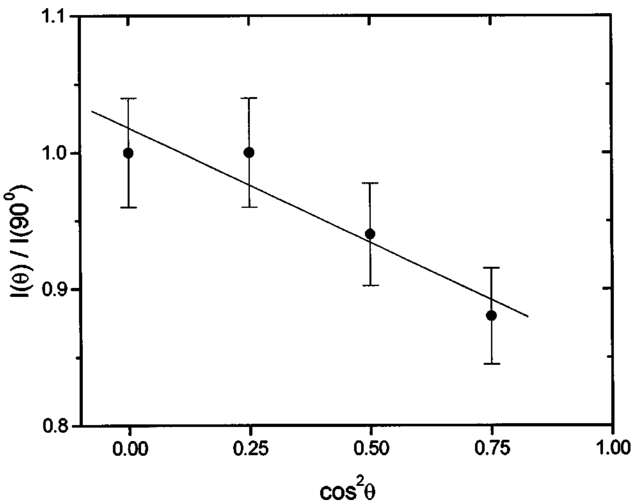

FIG. 4. Variation of the relative intensity $I(\theta)/I(90°)$ of argon K radiation at a given angle of emission θ, as a function of $\cos^2\theta$.

in Fig. 3, it is noted that the multiple-ionization reactions in argon might very likely take place through a simultaneous K- and multiple L-shell vacancy creations leading to a charge state of $+3$ and even more. When a simultaneous K- and multiple L-shell vacancies are created in the target atom, an unequal population of the magnetic substates in the $L_{2,3}$ shell is expected, that is, different populations in the magnetic substates of the $2\,^2P_{3/2}$ term should occur. Thus, a nonuniform distribution of spatial charge distribution in the $2\,^2P_{3/2}$ magnetic substates is expected to be manifest in the K-shell radiation emitted as a result of filling of K vacancy by electrons which are originating from a nonisotropic state $2\,^2P_{3/2}$ state.

IV. RESULTS AND DISCUSSION

The anisotropy of K radiation of argon is determined by plotting a graph for a relative intensity $I(\theta)/I(90°)$ at a given angle of emission θ, normalized to the intensity measured at angle $\theta=90°$, as a function of $\cos^2\theta$ [see Eq. (1)]. The ''scattering volume'' is formed by overlap of the electron beam ($\Phi=3$ mm) and the gas beam effusing from the multicapillary tube ($\Phi=5$ mm). This volume is not strictly a ''point'' scattering center. It was, therefore, necessary to apply a $\sin\theta$ correction factor to the data of all angular distributions. After taking into account this correction, the data were plotted for $I(\theta)/I(90°)$ versus $\cos^2\theta$, for the given incident electron energy. Such a plot is shown in Fig. 4 for 10 keV e^--Ar collisions. A polarization fraction $P=(17\pm6)\%$ was extracted from the graph. In all experiments, the value of $I(\theta)/I(90°)$ was found to be maximum around 90°. According to the relation [22], the polarization fraction P can be expressed in terms of subshell ionization cross sections σ_0 and σ_1, respectively, for $M_L=0$ and ±1 as

$$P=\frac{\sigma_0-\sigma_1}{\sigma_0+\sigma_1}. \tag{8}$$

The P will assume a positive value when σ_0 is greater than σ_1. That is, in the case of $2p$ subshells, the probability of excitation or ionization of $M_L=0$ is greater than the probability of excitation of the $|M_L|=1$ state. This situation can be explained in terms of the impact-parameter-dependent excitation or ionization function. In the present experiments, the range of b is selectively small as we require $1s$-shell ionization simultaneously with $2p$-subshell ionization. A simple estimate of the range of b yields values from 0.05 to 0.14 Å for the change of scattering angle θ from 60° to 20°, respectively. Further, the radii of the K and L shells of argon are determined to be 0.03 and 0.12 Å, respectively. These estimates suggest that the charge distribution of the $M_L=0$ substate of the $2p$ wave function which is concentrated along the beam direction (z axis) will have a large probability of overlap with the incident beam compared to that with the $|M_L|=1$ substate, for which the charge distribution is spread over large impact parameters. This qualitative picture describes the polarization results observed in our experiments for all incident energies of the present investigation.

Furthermore, Löw *et al.* [9] derived an approximate expression for the ratio of cross sections for multiple ionization of the L shell to single ionization of the K shell as

$$\sigma(KL^n)/\sigma(K)=a_n+b_n(\ln E)^{-1}+\cdots, \tag{9}$$

where a_n and b_n are impact-energy-independent terms, $E=E_0/E(KL_{2,3})$, E_0 is the impact energy, $E(KL_{2,3})$ is the energy to ionize in the K and L shells. It is noted from this equation that in addition to the shake-off probability given by the first term, there is an impact-energy-dependent contribution (given by the second term) to the multiple-vacancy production probability. Although the shake-off process following a K-shell ionization cannot create a nonzero alignment of the double-vacancy state, e.g., $KL_{23}\,^1P,\,^3p$ in the present case. However, there might be a considerable contribution to the $KL_{2,3}$ ionization from the presence of direct collisions between the primary electron and an electron in the K shell or a direct collision of the ejected K-shell electron with the L-shell electron, or from autoionization of $L_{2,3}$-shell electrons during the collision, which very well may lead to a noticeable alignment of the $^{1,3}P$ states. In the present experiments, as the range as well as the impact energy of the incident electrons are relatively large compared to those used in Ref. [11], the above-mentioned additional processes of $KL_{2,3}$ ionization may become important, leading to a considerable net linear polarization of the Ar $K\alpha$ x-ray line.

In addition, we have plotted a curve showing the variation of net polarization P of the K radiation of argon with impact energy in the range of 10.0 to 24.0 keV (see Fig. 5). The polarization is found to show a nearly constant value for all incident energies within the experimental uncertainty. This result suggests that, once the $KL_{2,3}$ double vacancy has attained a polarization of value $P=(17\pm6\%)$, the corresponding final charge state fraction of argon ions becomes almost constant with impact energy. We have, in fact, observed a constant charge state fraction of about 2% for the final triple vacancy in argon for electron-impact energies ranging from 10 to 24 keV in our recent work on ejected electron-ion

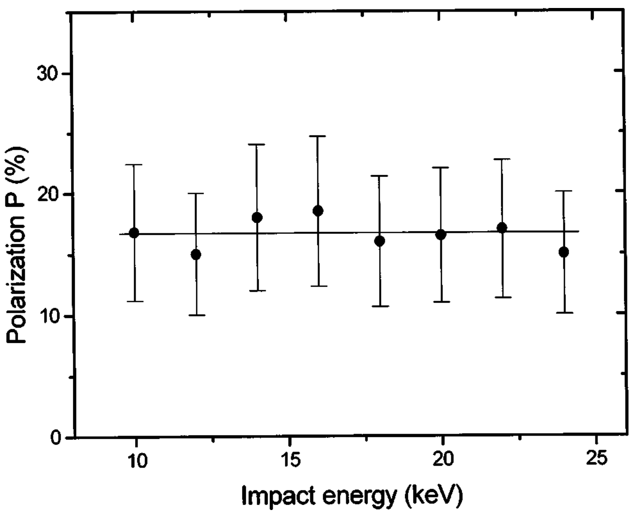

FIG. 5. Variation of the net polarization of K radiation of argon as a function of impact energy in the range of 10.0 to 24.0 keV. The solid line is a least-squares fit to the data points.

coincidence measurements (see Ref. [21]). This trend of variation of P with impact energy is, however, not in line with what the authors in Ref. [23] have reported for the degree of polarization P of a "thick" Al $K\alpha'$ ($^1P_1$-$^1S_0$) radiation. They found the degree of polarization to decrease with increasing impact energy when the target was bombarded by 1–3 MeV/amu He and Li ions. The different behaviors of polarization with impact energy in two observations may be explained by consideration of the two completely different collision conditions. In the case of a solid thick target, which was used by the authors of Refs. [7], [23], multiple collisions are unavoidable; these are expected to "smear out" the polarization degree in the end channel of x-ray detection especially at high impact energies; the K-shell photons for such collisions will be produced at a deeper location in the target and suffer a greater smearing effect at the detection channel compared to those photons that are produced by low-velocity ions. In contrast, our experiments are performed in a single-collision condition where the degree of polarization does not suffer from multiple collision effects.

V. CONCLUSIONS

In conclusion, our experimental data show a net linear polarization of $(17\pm6)\%$ of the K radiation of *free* gaseous target atoms of argon in bombardments with 10.0–24.0 keV electrons. This noticeable polarization is believed to arise from a strong polarization of $K\alpha$ satellite lines produced by the simultaneous decay of a single K and multiple L shells of argon atoms in a collision reaction. Also, our data exhibit a nearly constant value of the net polarization over the impact energies under consideration.

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