# Distribution of the number of emitted electrons for MeV $H^+$- and $He^{2+}$-ion impact on metals

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The statistical distribution of the number of emitted electrons induced by MeV $H^+$ and $He^{2+}$ impact on aluminum, copper, and gold targets was measured. The obtained results are very well represented by a Pólya distribution. Based on a simplified theoretical picture the appearance of a Pólya distribution in this context can be explained by cascade processes, which permits a quantitative estimation of the distributions in good agreement with the experiments for all investigated projectile-target combinations. The deviation from a Poisson distribution is given by the relative importance of cascade processes compared to projectile-induced processes.

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Ion-induced electron emission has been investigated for a long time and is important for many applications of ion impact on solids. Various aspects of ion-induced kinetic electron emission have been recently reviewed by Hasselkamp [1] and by Rothard, Groeneveld, and Kemmler [2]. Most of the reported investigations studied the total electron yield $\gamma$, which is the mean number of emitted electrons per impinging ion. The distribution of the number of emitted electrons has obtained less attention. Hofer [3] considered the statistics of electron emission in his review in more detail.

Up to now emission statistics were mainly measured for ion energies below 100 keV. Dietz and Sheffield [4] studied alkali-metal-ion impact on oxide films, and they found distributions which can be well described by Pólya distribution, whereas Thum and Hofer [5] found for stainless steel targets and heavy-ion impact Poisson distributions. Lakitis, Aumayr, and Winter [6] studied low-energy ion impact on Au, and they found neither a Poisson nor a Pólya distribution. Ohya, Aumayr, and Winter [7] explained their measured deviations from the Poisson distribution by large-angle scattering of incident ions and by the recoiling of target atoms. Azuma *et al.* [8] reported recently measurements of the number distribution of emitted electrons from C foils using He, C, and O ions of 1 MeV/u. They got distributions wider than Poisson distributions and they point out that cascade electrons may cause this broadening, but they did not compare their results with other distributions. Therefore up to now it is not clear which distribution describes the statistics of kinetic electron emission for MeV light-ion impact on metals and what physical processes determine the distribution.

In this paper we report systematic measurements of the distribution of emitted electrons for impact of $H^+$ and $He^{2+}$ ions with energies between 0.5 and 4.8 MeV on Al, Cu, and Au targets. The experimental setup was recently described in detail [9]. $H^+$ and $He^{2+}$ projectiles were obtained from the 1.6-MV tandem accelerator at Linz university. Measurements were performed in an ultrahigh-vacuum (UHV) vacuum chamber. The working pressure was $4\times10^{-10}$ mbar with all valves open to the beamline. A differentially pumped beam entrance chamber was used to separate the UHV chamber from the beamline vacuum system. The targets (Al, Cu, Au) were produced by vacuum evaporation on polished stainless steel backings and after preparation they were moved to a manipulator in the UHV chamber without breaking the vacuum. Before measurement, all targets were analyzed by Auger electron spectroscopy and they were sputter cleaned using 2-keV Ar ions until no carbon contamination was visible in the Auger spectrum. The angle of incidence of the ion beam with respect to the surface normal of the target was 21°. The targets were in the center of a cylinder opposite to a cylinder surrounding a solid-state detector which was on ground potential. The target and the target cylinder were at a potential $U=-20$ kV so that all the emitted electrons, up to an electron energy of 60 eV, were accelerated and focused to the detector. The electronic resolution was about 6 keV full width at half maximum (FWHM). After applying the high voltage and after conditioning, a background count rate of 20 $s^{-1}$ was reached which was negligible compared to the total count rate at 2000 $s^{-1}$.

The evaluation of the measured spectra was performed by a method similar to that used by Lakits, Aumayr, and Winter [10]; it is described in Ref. [9]. If $n$ electrons are emitted per impinging ion, they will produce a single pulse at the detector corresponding to an energy $neU$, if the total energy of the accelerated electrons is deposited in the detector. In the evaluation it is assumed that each individual electron with energy $E_0=eU$ has a probability $p$ to be reflected from the detector and deposits only part of the energy, $(1-k)E_0$, in the detector. Simultaneous emission of $n$ electrons therefore produces not one peak but an energy spectrum $F_n(E)$ which is the sum of $n+1$ peaks corresponding to the reflection of $m$ electrons $(0\le m\le n)$. The measured spectrum $S(E)$ is therefore fitted by

$$S(E)=\sum_{n=1}^{n_{\max}} C_n F_n(E) , \tag{1}$$

where $C_n$ gives the total number of events where $n$ elec-

 

trons were emitted. We use a fit program in which each of the parameters $p$, $k$, and $C_n$ can be either fitted or fixed to a constant value. For comparing the parameters $C_n$ with some theoretical distribution, any subset of $C_n$ can be bound to the distribution. Poisson, Gaussian, and Pólya distributions can be selected. We find that for all measured spectra the Pólya distribution [11]

$$P_n(\mu,b)=\frac{\mu^n}{n!}(1+b\mu)^{-n-1/b}\prod_{i=1}^{n}[1+(i-1)b] \qquad (2)$$

gives the best results, where $\mu$ is the mean value (total emission yields $\gamma$) and $b$ describes the deviation from a Poisson distribution (for $b=0$ the Pólya distribution becomes a Poisson distribution). Figure 1 shows a measured and a fitted spectrum, where the $C_n$ are bound to a Pólya distribution, for 1-MeV $H^+$ on Au. The reliability of the method was proved by comparing $\mu$ to the total emission yield $\gamma$ obtained by current integration [12]. For $\gamma \geq 4$, where $C_0$, which cannot be measured directly, is negligibly small, agreement was found within 2%.

All the measured spectra were then fitted with a Pólya distribution. Figure 2 gives the resulting $b$ values as a function of the projectile energy. For one type of projectile the order of low to high $b$ values is always Cu, Au, and Al, whereas the order of low to high $\gamma$ values is Al, Cu, and Au. Hence $b$ is no obvious function of $\gamma$. In the following a theoretical analysis of these experimental results will be given on the basis of a simplified model description.

As the impinging ion travels through the target surface layer (of typically about 5 nm depth) it gives energy kicks to the electrons surrounding its path. Let us assume that this process can be roughly subdivided into $N$ equivalent time steps of width $\Delta t$, each having a constant probability $p_p$ for the excitation of a bound electron by the projectile. Then the probability $P_n$ that altogether exactly $n$ electrons are liberated in $N$ "trials" (i.e., subsequent time steps $\Delta t$) is given by a binomial distribution, which reduces to a Poisson distribution, since $N\rightarrow\infty$ and

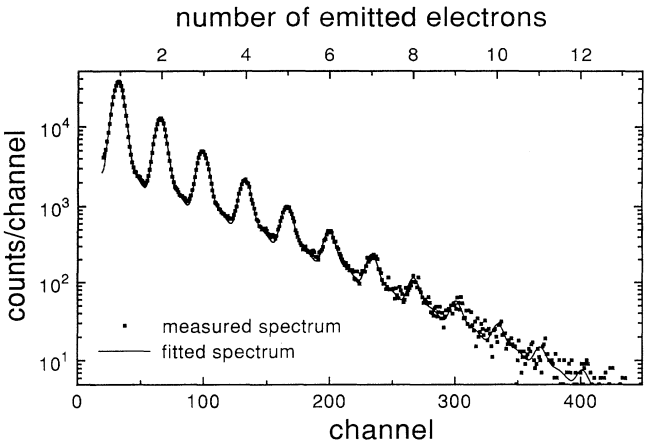


FIG. 1. Measured energy spectrum of electrons for 1-MeV proton impact on Au (dots). The full line corresponds to the fitted spectrum, assuming a Pólya distribution.

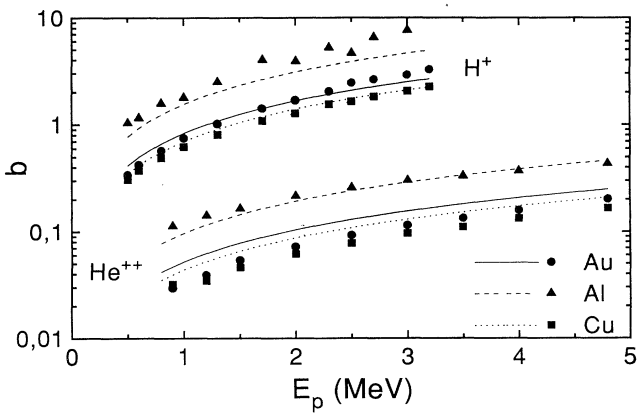


FIG. 2. Pólya parameter $b$ versus projectile energy for various projectile-target combinations. The lines are the results of the theoretical calculations using Eqs. (8) and (10).

$p_p\rightarrow 0$ (i.e., $\Delta t\rightarrow 0$) is the natural limit for the sampling procedure. This result also holds if $n$ now denotes the number of secondary electrons really *escaping* the target, since taking into account the processes of reaching the surface and surmounting its barrier merely leads to a redefinition of $p_p$ within this simplified picture.

However, as already discussed, the measured statistics significantly deviates from a Poisson distribution, especially for proton-induced electron emission. Consequently, an additional mechanism must be effective, which can be identified as the electron cascade: let us assume that each excited electron has an (averaged) constant probability $p_e$ of exciting another electron within each time interval $\Delta t$. The total chance $p_i$ for the $i$th electron to be liberated can then be estimated as

$$p_i=p_p+(i-1)p_e \ , \qquad (3)$$

provided that $p_p \ll 1$ and $p_e \ll 1$. With these assumptions $p_i \ll 1$ will also hold, and the exact distribution for the total excitation probability can be replaced by

$$P_n \propto \binom{N}{n}\prod_{i=1}^{n} p_i \ . \qquad (4)$$

In the limit $N\rightarrow\infty$ and $p_i\rightarrow 0$ Eq. (4) reduces to a Pólya distribution [Eq. (2)] $P_n(\mu,b)$ with parameter $b$

$$b=\frac{p_e}{p_p} \qquad (5)$$

and mean value $\mu$

$$\mu=\frac{Np_p}{1-Np_e}\approx n_p+n_p n_e \ , \qquad (6)$$

where we have introduced the symbols $n_p$ and $n_e$ for the average numbers of electrons "produced" by the projectile and one particular electron, respectively. As indicated by Eq. (6) this model is limited to the case $n_e \ll 1$, which is consistent with the assumptions leading to Eq. (3).

Again the escape process can be reintroduced easily

within this picture by redefinition of $p_p$ and $p_e$. We then can identify $\mu$ with the total emission yield $\gamma$ that, according to Eq. (6), consists of a sum of *direct* (projectile-induced) and *indirect* (electron-induced) contributions. Finally, Eq. (5) provides an interpretation of the Pólya parameter $b$ as a measure for the *cascade strength,* making $b$ accessible to theoretical analysis. This will be done in the following by a rough semiempirical model. In order to simplify the problem as much as possible we are making the following basic assumptions.

(i) The energy distribution of the electrons in the target can be replaced by a monoenergetic one, i.e., all electrons have approximately the same energy $\bar{\varepsilon}$. As a consequence $p_e$ is a constant and the steps leading to Eqs. (4)–(6) are still valid.

(ii) We neglect depth profiles, i.e., $p_p$ and $p_e$ are constant over the whole surface layer.

(iii) The probability distribution of energy kicks $\Delta E$ induced by a projectile (electron or ion) can be approximated by

$$f(\Delta E)=\frac{\pi}{\Delta E^2}\frac{Z^2e^4M}{m_eE_0}\ , \tag{7}$$

where $Ze$ and $M$ are the projectile charge and mass, respectively; $E_0$ is the projectile's kinetic energy. Integrating $-\Delta E f(\Delta E)$ from the mean ionization energy $I$ [13] (i.e., the minimum energy for exciting an electron, $E_{\min}$) to the maximum energy transferred in a head-on collision ($E_{\max}$) just leads to the close-collision contribution of the Bethe stopping formula. For more details, see Ref. [14].

(iv) We assume that—due to scattering processes—the escape probabilities for *directly* and *indirectly* excited electrons are the same. Thus the numerator and denominator in Eq. (5) are scaled with the same factor whose actual value is then irrelevant for the calculation of $b$.

As a next step, $p_e$ and $p_p$ are assumed proportional to the integral of $f(\Delta E)$ from Eq. (7) between $E_{\min}$ and $E_{\max}$. Using this in combination with Eq. (7) in Eq. (5), and with $E_{\min}\ll E_{\max}$, immediately yields

$$b\cong\frac{E_p}{\bar{\varepsilon}}\frac{m_e}{M_pZ_p^2}\ . \tag{8}$$

Here, $E_p$ and $M_p$ are the projectile (i.e., ion) kinetic energy and mass, respectively, and $Z_p$ denotes the ion (effective) charge [15] in unit charges; $m_e$ is the electron mass and $\bar{\varepsilon}$ the mean electron energy [see assumption (i)]. Thus the projectile's charge state appears squared in Eq. (8), which is characteristic for kinetic collision processes where wake-field effects have been neglected [16].

Comparing the measured values of $b(E_p)$ with Eq. (8) shows that $\bar{\varepsilon}$ depends on the projectile properties (energy, mass, charge) only very weakly. To a certain extent this is a surprising result, since it implies that nearly all information on the impinging projectile is lost during the cascade process. From another point of view, however, it appears to be somehow related to the well-known semiempirical model by Sternglass [17] for the secondary-electron *yield:* the quotient of yield and stopping power is approximately 0.1 Å/eV for all projectile-target combinations. Obviously, the randomizing multiple-scattering (and cascade) processes inside the target surface layer wipe out more detailed information on the triggering process (the projectile), so that only the amount of deposited energy remains relevant for the total emission yield.

Bearing in mind that the mean electron energy $\bar{\varepsilon}$ reflects the secondary-electron energy distribution inside the target surface layer, it is even possible to give a rough quantitative estimation of this parameter. Fitting Eq. (8) to the experimental results, it is seen that for all investigated targets $\bar{\varepsilon}>I$ holds, where $I$ is the mean ionization potential. Consequently, the mean number of cascade electrons excited by an electron of energy $\bar{\varepsilon}$ should be roughly given by $\bar{\varepsilon}/I$. On the other hand, this number can be expressed in terms of the electron's trajectory in the sample, too. Let $R(\bar{\varepsilon})$ be the average range an electron of energy $\bar{\varepsilon}$ will travel in the surface layer, and $\lambda(\bar{\varepsilon})$ be the mean free path (MFP) for inelastic (ionizing) scattering processes; then $R/\lambda$ should approximately equal $\bar{\varepsilon}/I$. For $R(E)$ we use the approximation [18]

$$R(E)\cong\frac{K}{\rho}E^{\gamma} \tag{9}$$

with

$$\gamma=1.68 \quad \text{and} \quad K=5.8\times10^{-3}\ \text{g cm}^{-3}(\text{eV})^{-\gamma}\,\text{Å}\ .$$

The density of the target material is denoted by $\rho$. In principle, the MFP $\lambda$ is a function of the electron energy that approximately follows a universal curve (i.e., is independent of the target material), as is seen for the total MFP [19]. The inelastic MFP is found to roughly equal to the elastic one in the energy range of consideration here ($\approx$300–800 eV), depending on the mechanisms built into the model [20]. Since the precise behavior of $\lambda(E)$ for ionizing collisions is difficult to estimate, and a more detailed analysis clearly lies beyond the scope of the present approximation, we make use of the fact that $\lambda(E)$ varies slowly compared to $R(E)$, and replace $\lambda(E)$ by an averaged value $\bar{\lambda}$.

Collecting all terms we finally obtain

$$\bar{\varepsilon}\approx\left[\frac{\bar{\lambda}\rho}{KI}\right]^{1/\gamma-1}\ , \tag{10}$$

where $K$ and $\gamma$ are defined in Eq. (9). From the behavior of MFP curves calculated from first principles, $\bar{\lambda}$ can be determined to be about 10–30 Å. Consequently, $\bar{\lambda}$ should be treated within this range of variation as a fit parameter that, nevertheless, must be fixed to a universal value independent of the target material (and, of course, the projectile properties, too).

Applying this model to our experiments on aluminum, copper, and gold, we find an extremely good agreement of the calculated with the measured Pólya parameters $b$ for all projectile-target combinations (see Fig. 2). We took $I$ and $\rho$ from the literature [13], and have set $\bar{\lambda}=19$ Å, leading to values of $\bar{\varepsilon}$ of 350, 780, and 655 eV for aluminum, copper, and gold, respectively. The projectile charge was set equal to the bare core charge, which should be a sufficient approximation for the present experiments, especially in the high-energy range [15].

Despite the tempting accuracy with which this estimation of $b$ agrees with our experiments, the reader should be aware that—to a certain extent—this does not necessarily prove the correctness of our assumptions. Although we believe that the steps leading to Eq. (8) reflect the dominating processes responsible for the emission statistics realistically enough, in particular, Eq. (10) for the parameter $\bar{\varepsilon}$ is meant as a high-energy ($>1$ keV) approximation, to use which at lower energies is at least questionable. Especially for gold, where $\bar{\varepsilon}/I$ (and thus $R/\lambda$) is even smaller than 1, the model obviously breaks down. However, this is a consequence of the oversimplifications that led to Eq. (10). In any case, our rough model is not meant to replace a more sophisticated analysis of the problem by first-principles calculations or simulations, but it provides a first approach to understanding the mechanisms responsible for our obtained experimental results.

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[1] D. Hasselkamp, in *Particles Induced Electron Emission II*, Springer Tracts in Modern Physics Vol. 123 (Springer, Berlin, 1992).
[2] H. Rothard, K. O. Groeneveld, and J. Kemmler, in *Particle Induced Electron Emission II* (Ref. [1]).
[3] W. Hofer, Scanning Microsc. Suppl. **4**, 265 (1990).
[4] L. Dietz and J. Sheffield, J. Appl. Phys. **46**, 4361 (1975).
[5] F. Thum and W. Hofer, Surf. Sci. **90**, 331 (1979).
[6] G. Lakits, F. Aumayr, and H. Winter, Phys. Lett. A **139**, 395 (1989).
[7] K. Ohya, F. Aumayr, and H. Winter, Phys. Rev. B **46**, 3101 (1992).
[8] T. Azuma, Y. Yamazaki, K. Komaki, M. Sekiguchi, T. Hasegawa, T. Hattori, and K. Kuroki, Nucl. Instrum. Methods Phys. Res. Sect. B **67**, 636 (1992); in *Ionization of Solids by Heavy Particles,* edited by R. Baragiola (Plenum, New York, 1993).
[9] O. Benka, E. Steinbauer, O. Bolik, and T. Fink, Nucl. Instrum. Methods Phys. Res. Sect. B **93**, 156 (1994).
[10] G. Lakits, F. Aumayr, and H. Winter, Rev. Sci. Instrum. **60**, 3151 (1989).
[11] W. Feller, *An Introduction to Probability Theory and its Applications* (Wiley, New York, 1957), Vol. 1, pp. 82ff.
[12] O. Benka, E. Steinbauer, and P. Bauer, Nucl. Instrum. Methods, B Phys. Res. Sect. B **90**, 64 (1994).
[13] S. P. Ahlen, Rev. Mod. Phys. **52**, 121 (1980).
[14] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1962), pp. 429ff.
[15] See Eq. (4.28) in Ref. [13].
[16] J. E. Borovsky and D. M. Suszcynsky, Phys. Rev. A **43**, 1433 (1991).
[17] E. J. Sternglass, Phys. Rev. **108**, 1 (1957).
[18] L. C. Feldman and J. W. Mayer, *Fundamentals of Surface and Thin Film Analysis* (North-Holland, New York, 1986), p. 143.
[19] See Ref. [18], Fig. 6.4.
[20] M. Rösler and W. Brauer, in *Particle Induced Electron Emission I,* Springer Tracts in Modern Physics Vol. 122 (Springer, Berlin, 1991).