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Information-theoretical interpretation of level populations and charge distribution in beam-foil spectroscopy

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The consequences of the assumption that, under prescribed experimental conditions, the ions emerge randomly excited from the foil are examined using the maximum entropy principle. For that purpose constraints are identified by examining the time-resolved intensity of the electromagnetic radiation emitted by the ions after the foil. This leads to the most likely level or multiplet population density matrix compatible with the experimental information. The main consequences of the foil excitation mechanisms are accounted for by incorporating the framework of atomic structure into our information-theoretical approach. In the case of transitions in individual Rydberg series it is shown that for large n and a given l the level population functions decrease as n^{*-3} and depend universally on the kinetic energy of the incoming beam. It is also shown that the identification of the radiation from the emerging ions in terms of one-electron states leads, according to the maximum entropy principle, to a unique factorized representation of the probability that an ion has a given configuration immediately after the foil. This representation is used for a derivation of the charge distribution which for heavy ions is shown to be approximately Gaussian or chi-squared in accordance with experiments. The passage of He^+ ions through the foil is treated as an example of a consistency test based on the information-theoretical derivation of the charge distribution. Following Levine and coworkers a surprisal analysis of level population functions is suggested as a well-established alternative to detailed models of foil excitation which usually invoke perturbation theory and consequently often have an ill-defined range of validity.

I. INTRODUCTION

In beam-foil spectroscopy relatively little is known about the excitation mechanisms in the foil.[1] This question is, however, relevant both for atomic lifetime experiments and for radiation physics in general, dealing particularly with the penetration of ions in condensed media.[2,3] In order to learn about the excitation mechanism one may study not only the characteristic radiation and electron emission pattern from the ions after the foil but also from the inside of the foil.[4] This paper deals solely with the analysis of post-foil measurements of the electromagnetic radiation and of the charge of the ions.

The foil acts as an abrupt excitation agent for the incoming ions and many processes like electron capture contribute to the final outcome. As a consequence of the low density of the beam (typically 10^5 ions/cm^3) the procedure of excitation which determines the level population of the ions immediately after the foil can be looked upon as an experiment of repeatedly exciting an ion into various states of variable charge ranging in principle from -1 to $+Z$. With this in mind we shall address ourselves to the general question of the possible information content in post-foil measurements regarding the excitation process. Our analysis is based on information theory[5] and on the application of the maximum entropy principle[6-8] to the derivation of the level populations and the charge distribution. This implies the assumption that the foil excites the ions completely at random to the available states which are subject to various constraints imposed by the experimental arrangement and by the atomic structure. As a result general predictions about the level populations and charge distribution are obtained. On the detailed level all experimental facts may not necessarily fit the information-theoretical distribution thus revealing dominant nonrandom features in the excitation process.

In Sec. II A we shall introduce the level-population density matrix ρ_p which is a function of the kinetic energy and charge of the incoming beam and which describes the distribution of the excited states in the ions immediately after the foil. The relationship[2,9,10] between ρ_p and the time-resolved intensity is examined in the case of cylindrical symmetry including the beat phenomenon. This leads to the identification of the constraints for the information-theoretical analysis of the level populations in Sec. II B. Our analysis shows explicitly how symmetry should be included in the constraints, a topic which has apparently not been discussed in the information-theoretical literature. In Sec. III A we apply our results to the characterization of the global behavior of level population functions of high Rydberg levels. A statistical derivation of the asymptotic level population is also given as an alternative to the usual arguments invoking perturbation theory.[11,12] In Sec. III B 1 we introduce a factorized representation for the classification of level populations. The probabil-

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ity that an ion emerges from the foil with a given configuration is shown to be a product of one-electron probabilities if and only if the maximum entropy principle is fulfilled. The factorized level population is used in Sec. III B 2 for a derivation of the initial charge distribution which is shown to be related in heavy ions to Gaussian or chi-squared distributions. It is also indicated how our results may be used to link experimental level populations to measured charge distribution in order to test the validity of the random-excitation hypothesis within the factorized-population framework. The passage of He^+ ions through the foil[11,13] is treated as an example by including the autoionization rate explicitly.

We shall conclude our paper with a brief discussion on the uniqueness of the maximum entropy principle with respect to predictions about the random-excitation hypothesis. On the practical level a surprisal analysis[14,15] of post-foil data is suggested. The population of Rydberg levels is treated as an example.

II. INFORMATION-THEORETICAL POST-FOIL ANALYSIS

A. The relationship between level population and observed intensity

We assume that the foil is perpendicular to a uniform ion beam and that there are no external fields. After suffering various inelastic collisions in the foil and its vicinity the ions usually emerge somewhat deflected and with less kinetic energy into the region of observation. They may have lost or gained electrons and emit electromagnetic radiation which is observed in a given direction and with a given polarization analyzer arrangement. By varying the distance from the foil a time-resolved intensity distribution is obtained.[2]

It is in principle impossible to separate the decay from the foil interaction. Nevertheless the high velocity of the ions makes it meaningful to define a *level population* density matrix $\rho_p(T,Q)$ which refers exclusively to the excitation by the foil. The matrix ρ_p depends on the kinetic energy T and charge Q of the incoming ion beam and determines at $t=0$ the initial condition for the density matrix $\rho(t)$ which describes the subsequent photon emission and autoionization including cascade effects. Consequently the matrix $\rho(t)$ satisfies a Liouville equation in which the Hamilton operator refers solely to the free ions, the radiation field and their interaction. If ρ_p is known, then $\rho(t)$ completely determines the time-resolved intensity distribution

$$I(t)=\mathrm{Tr}\rho(t)F, \tag{1}$$

where F is the matrix of the detector operator $\hat{F}$ (Ref. 16) which is specified by the experimental arrangement.

For electric dipole transitions and an ideal polarization sensitive detector $\hat{F}$ is given by

$$\hat{F}=(\vec{\epsilon}\cdot\vec{r}')\hat{P}_f(\vec{r}',\vec{r})(\vec{\epsilon}^{\,*}\cdot\vec{r}), \tag{2}$$

where $\vec{\epsilon}$ is a (complex) polarization vector and $\hat{P}_f(\vec{r}',\vec{r})$ the projection operator pertaining to all the unobservable final states.[9] Since the measurements do not resolve the magnetic substates, $\hat{P}_f$ includes summations over complete sets of magnetic quantum numbers and, as a consequence, it is a scalar operator. This property allows $I(t)$ to be expressed, after a recoupling procedure, as the weighted sum

$$I(t)=\sum_{K=0}^{2}\sum_{QQ'}(-1)^{K+Q}\mathcal{S}^{(K)}_{-Q}(\vec{\epsilon},\vec{\epsilon}^{\,*})D^{(K)}_{Q'Q}(\omega)\times\mathrm{Tr}[\rho(t)R^{(K)}_{Q'}(\vec{r}',P_f\vec{r})] \tag{3}$$

of the polarization factors[17]

$$\mathcal{S}^{(K)}_{-Q}(\vec{\epsilon},\vec{\epsilon}^{\,*})=\sum_{qq'}\epsilon_q\epsilon^{*}_{q'}\langle 11qq'|K-Q\rangle. \tag{4}$$

The matrix elements, $D^{(K)}_{Q'Q}(\omega)$, of finite rotations transform the dynamical tensor components $\hat{R}^{(K)}_{Q'}$ from the collision frame with the z axis parallel to the beam axis into the detector frame with the z axis parallel to the direction of observation. The weight factors $\mathrm{Tr}[\rho(t)R^{(K)}_{Q'}(\vec{r}',P_f\vec{r})]$ can be further simplified by explicitly restricting the trace to the initial states Γ involved in the observed transition and by taking the eventual symmetry of $\rho(t)$ with respect to the collision frame into account.

In the following we shall consider two limiting cases of Eq. (3), namely a transition $\gamma^i_q J_i-\gamma^f_q J_f$ between two levels and a multiplet $\gamma^i_q S_i L_i-\gamma^f_q S_f L_f$. Here γ_q refers to the configuration of an ion with charge q and to any quantum numbers which may be needed to describe a level or multiplet in addition to the angular momentum quantum numbers. We shall neglect for the present purposes the hyperfine interaction so that $\rho(t)$ is given either with respect to a $\{|\gamma_q JM\rangle\}$ or $\{|\gamma_q SLM_S M_L\rangle\}$ representation in the collision frame.

It is usually assumed that in the perpendicular situation the ion beam is cylindrically symmetric after the foil. This assumption which is by no means self-evident simplifies Eq. (3) considerably.[10]

Cylindrical symmetry requires that $\rho_{\gamma'_q J'M',\gamma_q JM}(t)$ is diagonal in M which implies that $Q'=0$ in Eq. (3). Furthermore it includes reflection symmetry which requires that the matrix elements of ρ de-

pend only on $|M|$, restricting K to even values. It is then a matter of angular momentum algebra to transform Eq. (3) into a convenient form[18] which for the $\gamma_q^i J_i \to \gamma_q^f J_f$ transition is given by

$$I=\frac{1}{2J_i+1}|\langle J_f\|\vec{r}\|J_i\rangle|^2 \mathrm{Tr}_\Gamma\rho(t)[1+kf(\theta,\psi,\beta)], \quad (5)$$

where Tr_Γ refers to a summation over M_i and where

$$f(\theta,\psi,\beta)=\tfrac{3}{2}\sin^2\theta\cos2\psi\cos2\beta-P_2(\cos\theta). \quad (6)$$

Here θ is the angle between the direction of observation and the beam axis. As described in detail in Refs. 9 and 19 the angles ψ and β account for the polarization selected by the detector. By using the Wigner-Eckart theorem the anisotropy factor k can be expressed as

$$k=\frac{(-1)^{J_i-J_f}\begin{Bmatrix}J_i & J_i & 2\\ 1 & 1 & J_f\end{Bmatrix}}{2J_i(J_i+1)\begin{Bmatrix}J_i & J_i & 2\\ 1 & 1 & J_i\end{Bmatrix}}\frac{\mathrm{Tr}_\Gamma[\rho(t)(3J_z^2-J^2)]}{\mathrm{Tr}_\Gamma\rho(t)}, \quad (7)$$

which displays the identical transformation properties of $\hat{R}_0^{(2)}$ and the angular momentum operator $3\hat{J}_z^2-\hat{J}^2$.[9]

Equation (5) implies that $\rho_{\gamma_q^i J_i,\gamma_q^i J_i}(t)$ stays cylindrically symmetric as the time evolves. This is even true if cascade effects are involved in the population of the level $\gamma_q^i J_i$.[10] Hence, for $t\gtrsim 0$ we may put[2] $\rho(t)=\rho_p\exp(-\Gamma_0 t)$, where Γ_0 is a diagonal matrix. This establishes the connection between ρ_p and a measurement of the intensity (5) at short distances from the foil.

Next we consider[20] the modification of Eq. (5) for a multiplet transition $\gamma_q^i S_i L_i \to \gamma_q^f S_f L_f$ with eventually an unresolved fine structure. The matrix $\rho(t)$ is now given in a $\{|\gamma_q SLM_S M_L\rangle\}$ representation. Since $\hat{R}_Q^{(K)}$ does not depend on the spin we may use the statistical tensors

$$\rho_Q^{(K)}(LL;t)=\sum_{M_L M_L'}\left(\sum_{M_S}\rho_{SM_S LM_L,SM_S LM_L'}(t)\right)\times(-1)^{L-M_L}\langle LM_L'L-M_L|KQ\rangle \quad (8)$$

to factorize the weight factors into

$$\mathrm{Tr}[\rho(t)R_Q^{(K)}]=\frac{\langle\gamma_q^i L_i\|R_Q^{(K)}\|\gamma_q^i L_i\rangle}{(2K+1)^{1/2}}\rho_{-Q}^{(K)}(L_iL_i;t). \quad (9)$$

Hence according to Eqs. (3) and (9) the statistical tensors (8) completely determine the time-resolved intensity of a multiplet. It also follows that in the absence of the spin-orbit interaction Eqs. (5)–(7) are valid for the multiplet intensity provided the replacements $L_i \to J_i$ and $L_f \to J_f$ are made everywhere.

Instead of the tensors (8) we may also form by an alternative coupling scheme the statistical tensors $\rho_Q^{(K)}(JJ';t)=\rho_Q^{(K)}((SL)J,(SL)J';t)$ with respect to the uncoupled representation $\{|\gamma_q SLM_S M_L\rangle\}$. The corresponding recoupling transformation is given by

$$\rho_Q^{(K)}(LL;t)=\sum_{JJ'}C_K(J'J;SL)\rho_Q^{(K)}(JJ';t), \quad (10a)$$

where

$$C_K(J'J;SL)=(-1)^{S+L+J+K}[(2J'+1)(2J+1)]^{1/2}\begin{Bmatrix}J' & J & K\\ L & L & S\end{Bmatrix}. \quad (10b)$$

As shown below these equations can be used for an approximate description of the modulation (the so-called beat phenomenon[2,10]) of the time-resolved intensity of a multiplet which has an unresolved fine structure due to the spin-orbit interaction.

The beat phenomenon is illustrated by the fact that

$$\rho_Q^{(K)}(JJ';t)=\rho_Q^{(K)}(JJ';0)\exp\left(\frac{i}{\hbar}(E_J-E_{J'})t\right), \quad (11)$$

with respect to a representation which diagonalizes the Hamiltonian matrix which includes the spin-orbit interaction. If this interaction is small, then the substitution of Eq. (11) into Eq. (10a) shows approximately how the $J\neq J'$ components of $\rho_Q^{(K)}(JJ';t)$ induces in the case of $K>0$ slow modulating oscillations of $\rho_Q^{(K)}(LL;t)$ and as a consequence of Eq. (9) also of the time-resolved multiplet intensity. Note that this requires that there is alignment, i.e., $\rho(t)$ has diagonal elements which depend on the magnetic quantum number.

Usually it is assumed that at $t=0$ the level population is independent of spin. Using the inverse of the transformation (10a) it can be shown that, as a consequence,

$$\rho_Q^{(K)}(JJ';0)=(2S+1)^{-1}C_K(JJ';SL)\rho_Q^{(K)}(LL;0),$$

where C_K is given by Eq. (10b). According to Eqs. (10a) and (11) this leads to a factorization of the statistical tensors (8) into

$$\rho_Q^{(K)}(LL;t)=\rho_Q^{(K)}(LL;0)D_K(t), \quad (12)$$

where the depolarization factor[21] is given by

$$D_K(t)=\sum_{JJ'}\frac{(2J+1)(2J'+1)}{(2S+1)}\begin{Bmatrix}J' & J & K\\ L & L & S\end{Bmatrix}^2\cos\omega_{JJ'}t. \quad (13)$$

In this case the multiplet intensity corresponding to Eq. (5) takes the form

$$I=\frac{1}{(2L_i+1)}\left|\langle L_f\|\tilde{\mathbf{r}}\|L_i\rangle\right|^2$$

$$\times \mathrm{Tr}_\Gamma\rho_\beta e^{-\Gamma_0 t}[1+kD_2(t)f(\theta,\psi,\beta)]\,, \qquad (14)$$

where k is given by Eq. (7) with $J_i\to L_i$, $J_f\to L_f$, and $\rho(t)\to\rho_\beta$.

Although the results (5) and (14) are rather well known in one form or another they have been re-examined for the purpose of the information-theoretical analysis of the level-population density matrix $\rho_\beta(T,Q)$. The important result is that in the cylindrically symmetric situation measurements of the time-resolved intensity $I(t)$ identifies at most two expectation values with respect to ρ_β, namely, $\mathrm{Tr}_\Gamma\rho_\beta$ and $\mathrm{Tr}_\Gamma\rho_\beta(3J_z^2-J^2)$ for each line observed. With regard to a test of cylindrical symmetry, note that the radiation may exhibit linear but not circular polarization.[10] The observation of multiplet transitions contains in general the same information but only with respect to L although the beat phenomenon may provide some insight in the possible spin dependence of ρ_β. However, as Eqs. (13) and (14) indicate the main effect can be attributed to the unresolved fine structure of the free ions *after* the foil.

B. Information-theoretical analysis of level populations

The excitation processes in the beam-foil interaction are poorly understood. In fact, even the theory of electron capture and loss in heavy ion-atom collisions is far from complete for single-collision conditions. Physical models of the presumable dominating beam-foil interaction mechanisms have been constructed for limited purposes like for the explanation of the initial level population of Rydberg series.[22] Detailed mechanisms have sometimes been combined with statistical arguments in order to parametrize initial-level populations and final charge distribution.[23,24] In the following we shall adopt an entirely different approach which is based on the maximum entropy principle in information theory[5-8] and consequently ignoring any details concerning the excitation mechanisms.

In the case of systems like the present one the information-theoretical strategy consists of a determination of the *most likely* or *probable* density matrix ρ for any selected excitations or fragments that are produced by the interaction. Such a matrix should not only be consistent with *random* excitation but also with the available information concerning the influence of the interaction on the residual system under observation. This is achieved by a maximization of the information-theoretical entropy

$$S_i=-\mathrm{Tr}(\rho\ln\rho) \qquad (15)$$

subject to constraints which are specified by the mean values

$$\langle\hat{T}_\mu\rangle=\mathrm{Tr}(\rho T_\mu)\,,\quad \mu=1,2,\ldots,M \qquad (16)$$

of a set of operators $\hat{T}_\mu$. The choice of the operators $\hat{T}_\mu$ is usually dictated by more or less implicit dependence of the measured quantities on the expectation values (16). In fact, the link between the measurements and the constraints (16) may also depend on various assumptions regarding the nature of the interaction, e.g., the separation of decay from excitation.

As it has been shown by Jaynes[7] and others, the operator

$$\hat{\rho}=\exp\left(-\sum_{\mu=0}^{M}\lambda_\mu\hat{T}_\mu\right) \qquad (17)$$

makes the entropy (15) maximum under the constraints (16). In Eq. (17) the Lagrange parameter λ_0 which corresponds to the identity operation accounts for the normalization $\mathrm{Tr}\rho=1$.

For the purpose of illustration of the maximum entropy principle consider its application to the blackbody problem. We make use of the fact that the distribution $P(\nu)$ for $\nu=0,1,2\ldots$, which maximizes $S_i=-\sum_{\nu=0}^{\infty}P(\nu)\ln P(\nu)$ subject to the constraints $\epsilon=\sum_{\nu=0}^{\infty}\nu P(\nu)$ and $\sum_{\nu=0}^{\infty}P(\nu)=1$ is given by $P(\nu)=e^{-\lambda\nu}/\vartheta$, where λ is the Lagrangian parameter and ϑ the "partition function" $\sum_{\nu=0}^{\infty}e^{-\lambda\nu}$.[25] Since $P(\nu)$ is known, the mean value ϵ can be related to λ, leading to $\epsilon=(e^{\lambda}-1)^{-1}$ which determines λ. The blackbody radiation law follows now from the identification of ν with $n\hbar\omega$ and from the definition of the temperature $T^{-1}\equiv\partial S/\partial\epsilon'$ and of the entropy by $dS\equiv kdS_i$.[6] Then $\lambda=\hbar\omega/kT$ and $\epsilon'=\epsilon\hbar\omega=\hbar\omega/(e^{\hbar\omega/kT}-1)$, which after the multiplication with the number of modes per unit volume gives Planck's radiation law. This result can be interpreted as a manifestation of the random character of the interaction between the radiation field and the wall. The only observable is the *mean* energy $\epsilon=\epsilon(\omega,T)$, related to the intensity distribution of the radiation from a small hole in any of the walls.

Returning to the determination of the level-population density matrix ρ_β from beam-foil experiments we note that the experimental arrangement corresponding to Eqs. (5), (7), and (14) yields information about

$$\mathrm{Tr}_\Gamma\rho(0)=\mathrm{Tr}\rho_\beta P_\Gamma=\langle\hat{P}\rangle_\Gamma\,, \qquad (18a)$$

$$\mathrm{Tr}\rho_\beta(3J_z^2-J^2)P_\Gamma=\langle 3\hat{J}_z^2-\hat{J}^2\rangle_\Gamma\,, \qquad (18b)$$

in the collision frame. In Eqs. (18) the projection operator

$$\hat{P}_\Gamma = \sum_{\kappa\in\Gamma} \hat{P}_\kappa = \sum_{\kappa\in\Gamma} |\kappa\rangle\langle\kappa| \tag{19}$$

corresponds to a summation over the states $|\gamma_q JM\rangle$ of an energy level or over the states $|\gamma_q SLM_S M_L\rangle$ of a multiplet in which case $J=L$ in Eqs. (18). In addition, the observation of lines or multiplets is supposed to identify the energy of the initial and final states and the number of electrons in the ions which emerge from the foil. This introduces the constraints

$$\mathrm{Tr}(\rho_p H P_\Gamma) = E_\Gamma\,, \tag{20a}$$

$$\mathrm{Tr}(\rho_p N) = \langle N_q\rangle\,. \tag{20b}$$

However, since E_Γ is an eigenvalue of the Hamilton operator $\hat{H}$ of the free ion the constraint (20a) reduces to $E_\Gamma\langle\hat{P}\rangle_\Gamma$ and hence adds nothing new to the constraints (18). The constraint (20b) in which N corresponds to the number operator $\hat{N}$ represents the available information about the $t=0$ charge distribution. Note that the constraints (18) and (20) contain information about *individual* stationary states under symmetry restrictions and should hence be contrasted with the blackbody case, where a *mean* energy is observed without an axis of preference.

From Eq. (17) it follows that

$$\hat{\rho}_p = \exp\left(-\lambda_0\hat{I} - \sum_\Gamma \lambda_\Gamma\hat{P}_\Gamma - \sum_\Gamma \lambda'_\Gamma(3\hat{J}_z^2 - \hat{J}^2)\hat{P}_\Gamma - \mu\hat{N}\right) \tag{21}$$

maximizes the entropy (15) subject to the constraints (18) and (20) given for each Γ observed. In Eq. (21) $\hat{I}$ is the identity operator. Since the operators appearing in the exponent of the operator (21) commute there exists a basis in which ρ_p is diagonal. This basis is given by $\{|\gamma_q JM\rangle\}$, respectively, $\{|\gamma_q SLM_S M_L\rangle\}$, leading then to

$$\rho_{\kappa'\kappa} = \delta_{\kappa'\kappa}\exp\{-\lambda_0 - \lambda_\Gamma - \lambda'_\Gamma[3M^2 - J(J+1)] - \mu N_q\} \tag{22a}$$

or

$$\rho_{\kappa'\kappa} = \delta_{\kappa'\kappa}\exp\{-\lambda_0 - \lambda_\Gamma - \lambda'_\Gamma[3M_L^2 - L(L+1)] - \mu N_q\}\,, \tag{22b}$$

where the Lagrangian parameters λ_Γ, λ'_Γ, and μ are functions of the kinetic energy T and charge Q of the incoming beam. Our derivation of the matrices (22) shows explicitly the consequence of symmetry constraints.

Lower symmetry implies an increase of the number of statistical tensors which in principle can be evaluated from the experiments. For instance, if reflection symmetry is not assumed in the perpendicular beam-foil case, then the intensity expression accounts for a net orientation along the beam axis which is described by $\hat{R}_0^{(1)}$ in Eq. (3). From an information-theoretical point of view this gives rise to a new constraint which makes the exponent in Eqs. (22) also dependent on the sign of M. Hence one may argue that the most general post-foil experiment yields information which is consistent with a maximum-entropy distribution of the form

$$\rho_{\kappa'\kappa} = \exp(-\lambda_{\kappa'\kappa} - \mu N_q)\,, \tag{23}$$

where also nondiagonal elements appear due to foil tilting.

Equation (22b) applies to the beat phenomenon[2,10] since it predicts that the states $|\gamma_q SLM_S M_L\rangle$ for a given S and L may be aligned. Note that the matrix elements (22b) are independent of M_S which is also a consequence of Eq. (14).

If the distributions (22) and (23) do not provide a basis for *global* predictions regarding the level populations and the related charge distribution, one has not achieved much more than a reparametrization of a limited set of experimental data. In order to achieve such predictions it is necessary to incorporate in the information-theoretical analysis of ρ_p specific consequences of the atomic structure. In the next section we shall make use of two: the concept of configurations and the density of Rydberg states. The information-theoretical framework does not rule out other possibilities.

III. IMPLICATIONS AND APPLICATIONS OF THE MAXIMUM ENTROPY PRINCIPLE

A. Level population function of Rydberg series

We shall begin with a derivation of the asymptotic level population function of Rydberg states using statistical arguments based on a modification of Feynman's[26] treatment of the Boltzmann distribution. This result is found to be in accordance with the maximum entropy principle provided normalization is the only constraint. The modification due to the constraints (18a) and (20b), necessary for identification of a series and the number of electrons, is made.

Usually the kinetic energy, T, of the incoming ions is much larger than any of the observed electronic excitation energies in the foil. Hence the total energy E_i of an outgoing ion can be anywhere within a given narrow energy interval $[E, E+dE]$ in which each state has an equal chance to become populated if random excitation is assumed. As a consequence the corresponding probability must be inversely proportional to the

number of states, $\eta(E_i)dE$, in the interval $(E, E+dE)$. In the case of a continuous or quasicontinuous spectrum $\eta(E)$ represents the density of states. This circumstance can be used to evaluate the probability P_{nl} that a high Rydberg state $n_2 l\ ^2L$ becomes populated relative to another state, corresponding to n_1 in the same series. It follows that

$$\frac{P_{n_2 l}}{P_{n_1 l}} = \frac{\eta(E_i')dE}{\eta(E_i'')dE} = \frac{\eta(T')\eta(E_{n_1 l})}{\eta(T'')\eta(E_{n_2 l})}, \tag{24}$$

where $E_i = T + E_{nl}$. In Eq. (24) $\eta(T') \cong \eta(T'')$ is the free-particle density of states associated with the outgoing nucleus and $\eta(E_{nl})$ the density of the states in a given Rydberg series. Since $E_{nl} = \frac{1}{2}\overline{Z} n_l^{*-2}$ in terms of the effective principal quantum number $n^* = n_l^*$, $\eta(E_{nl})$ is given by

$$\eta(E_{nl}) = \frac{dn^*}{dE_{nl}} = \frac{n^{*3}}{\overline{Z}}. \tag{25}$$

From Eqs. (24) and (25) it follows that for large n the *relative* probability P_{nl} should fulfill the relationship

$$\ln P_{nl} = -3 \ln n^* + C, \tag{26}$$

where C is a constant.

The result (26) is in accordance with the maximum entropy principle for continuous distributions.[7] It is required that

$$S_I = -\int_\Delta p(E) \ln \frac{p(E)}{\eta(E)} dE \tag{27}$$

is a maximum for a given energy interval Δ. In Eq. (27) $p(E)$ is the probability per unit energy that an excitation occurs into the interval $(E, E+dE)$. With the normalization requirement as the only constraint it can be shown that $p(E) = \eta(E)$ maximizes the entropy (27).[27] Hence the asymptotic behavior (26) of P_{nl} reflects the random excitation of Rydberg states also from the point of view of information theory.

So far the fact that the level population function measurements identify specific Rydberg series in given ions has not been considered. Usually alignment is lost in these experiments which are carried out with a polarization insensitive detector fixed at a given angle to the outgoing beam.[2] Hence we shall put $\lambda_\Gamma' = 0$ in Eqs. (22) and modify these distributions in accordance with Eq. (26). The result can obviously be written as

$$\rho_{\kappa'\kappa} = \delta_{\kappa'\kappa} \exp(-\lambda_0 - 3 \ln n^* - \lambda_l - \mu N_e), \tag{28}$$

where we have neglected the spin-orbit interaction so that $\kappa = (nlm l m_s)$. The parameter λ_l in Eq. (28) accounts for the identification of a given Rydberg series.

Equation (28) implies in accordance with Eq. (26) that

$$\frac{P_{n_2 l}}{P_{n_1 l}} = \left(\frac{n_1^*}{n_2^*}\right)^3 \tag{29}$$

and that $\lambda_l = \lambda_l(T)$ is a function of T characterizing the whole Rydberg series. Note that in Eq. (28) μ depends for a given incoming ion on T only.

The asymptotic behavior (26) or (29) of the relative Rydberg state population P_{nl} is observed frequently[11-13,28] but not exclusively.[29] Slight deviations[11-13,28] from the n^{*-3} law at low n can be attributed to the breakdown of the concept (25) of density of states. Large deviations would indicate that there is a single dominant process which populates the Rydberg levels. Evidence of such a process has recently been found by Andresen *et al.*[29] Their experiments indicate that in the $n = 3$ to 12 range $n \simeq 7$ and $\simeq 8$ are predominantly populated among the Rydberg states of Li-like O^{5+} and F^{6+} ions which have traversed carbon foils. They attribute this effect to resonant electron capture from the valence band of carbon. Numerous experiments[13,28,30] also indicate that for several Rydberg series in various ions the level population function is independent of T over a wide range of n in accordance with Eq. (28). As indicated above this behavior is also predicted by the maximum entropy principle and thus is in accordance with the assumption of randomly excited Rydberg states.

The validity of Eq. (26) is often taken as a verification of electron capture from the back of the foil into a high Rydberg state of the emerging ion.[12,24,31] The reason for this is that for high n *and* asymptotically in T the ratio of the Born cross sections is also determined by Eq. (29) through the amplitude $\eta(E_{nl})^{-1/2}$ of the Rydberg-state wave function. However, as shown above the same conclusion follows from more general arguments of statistical nature which are also accordance with the maximum-entropy principle. In addition, information theory predicts a universal behavior of the level populations as a function of T in the domain, where Eq. (28) is valid.

B. Relationship between level populations and charge distribution

1. The factorized representation

We would like to characterize the beam-foil measurements in terms of one-electron states. As a consequence, the level population function $\rho_\beta(T, Q)$ is described in terms of configurations

rather than energy levels of multiplets. The spectral data and the independently obtained charge distribution provide limited information from which we may extract the probabilities Q_ν that a one-electron state ν is occupied in an ion after the foil. The mean number of electrons $\langle N_q \rangle$ may also be obtained from the final charge distribution if the autoionization rates are known. Within the framework of information theory one can derive the most likely probability distribution $\{p_\kappa\}$ of the configurations and consequently also of the charge distribution immediately after the foil.

The probabilities $\{p_\kappa\}$ may be obtained from the maximum-entropy principle by the introduction of the constraints

$$\sum_\kappa p_\kappa = 1 , \tag{30a}$$

$$\sum_\kappa{}' p_\kappa = Q_\nu , \tag{30b}$$

$$\sum_{\kappa'} p_{\kappa'} N_{\kappa'} = \langle N_q \rangle . \tag{30c}$$

In the sum (30b) only those configurations κ with N_κ electrons are included that have an occupied one-electron state ν. Note that the configurations κ should run over all possible excitations and charges. Following Bard[32] it can be shown that the maximum of the entropy (15) subject to the constraints (30) is uniquely given by the distribution

$$p_\kappa = \frac{\exp(-\sum_{\nu\in\kappa}\lambda_\nu - \mu N_\kappa)}{1+\sum_{\kappa\neq 0}\exp(-\sum_{\nu\in\kappa}\lambda_\nu - \mu N_\kappa)} , \tag{31}$$

where the denominator can be identified as the partition function ϑ. Note that the bare-ion probability p_0 is normalized to ϑ^{-1}. From Eq. (31) it follows that p_κ is proportional to a *product* of probabilities $\exp[-\lambda_\nu - \mu]$ establishing the occupancy of a one-electron state. Note that in this factorized representation there is *no* specification of the one-electron model used in the construction of the configurations. In analogy with a work of Abdulnur *et al.*[33] the result (31) can be expressed in the concise form

$$p_\kappa = \frac{\langle \kappa | \hat{\rho} | \kappa \rangle}{\sum_\kappa \langle \kappa | \hat{\rho} | \kappa \rangle} \tag{32}$$

by using the density operator

$$\hat{\rho} = \exp\left(\sum_\nu \lambda_\nu \hat{n}_\nu - \mu \sum_\nu \hat{n}_\nu\right) = \prod_\nu (1+\alpha_\nu \hat{n}_\nu) , \tag{33}$$

where $\hat{n}_\nu = a_\nu^\dagger a_\nu$ is the occupation number operator of state ν and where

$$\alpha_\nu = e^{-\lambda_\nu - \mu} - 1 . \tag{34}$$

If the configurations κ would include all possible combinations $\{n_1, n_2, \ldots, n_\nu, \ldots\}$ with occupation numbers n_ν zero or one, without any limitations on maximum N_κ, the each p_κ is given by

$$p_\kappa = \prod_{\nu=1}^{N_\kappa} \frac{(1+\alpha_\nu)}{(2+\alpha_\nu)} = \prod_{\nu=1}^{N_\kappa} \langle \hat{n}_\nu \rangle \tag{35}$$

according to Eqs. (32) and (33). According to Eq. (34) the average occupation number $\langle \hat{n}_\nu \rangle$ of state ν may also be written as

$$\langle \hat{n}_\nu \rangle = [1+\exp(\lambda_\nu + \mu)]^{-1} , \tag{36}$$

which would reduce to the Fermi distribution law if a "temperature" parameter θ could be defined such that $\lambda_\nu = \epsilon_\nu/\theta$ for each ν. This is not in general the case for foil excitation.

2. *Charge distribution*

We shall now use Eq. (31) to establish the connection between level populations and the initial charge distribution defined by

$$P_{N_q} = \sum_{\kappa\in q} p_\kappa , \quad q = -1, 0, \ldots, +Z \tag{37}$$

where q indicates that all configurations corresponding to a definite number, N_q, of electrons should be included in the sum. The upper limit of q may not always refer to the bare nucleus in the following. Information about the post-foil occupancy of inner-shell states usually requires x-ray measurements. Hence Z should be understood to be the nuclear charge eventually screened by a given inner-shell core.

With the notation $x_\nu = \exp(-\lambda_\nu - \mu)$ we obtain

$$\begin{aligned}
P_0 &= \vartheta^{-1} , \\
P_1 &= \vartheta^{-1} \sum_{\nu=1}^{M} x_\nu , \\
P_2 &= \vartheta^{-1} \sum_{\nu_1>\nu_2}^{M} x_{\nu_1} x_{\nu_2} , \\
&\cdots \\
P_k &= \vartheta^{-1} \sum_{\nu_1>\nu_2>\cdots>\nu_k}^{M} x_{\nu_1} x_{\nu_2} \cdots x_{\nu_k} , \\
&\cdots \\
P_{N+1} &= \vartheta^{-1} \sum_{\nu_1>\nu_2>\cdots>\nu_{N+1}}^{M} x_{\nu_1} x_{\nu_2} \cdots x_{\nu_N} ,
\end{aligned} \tag{38}$$

where M is the number of one-electron states included in the analysis. Usually M can be taken to be very large due to the large energy of the incoming beam. Note that antisymmetry requires that a given x_ν can occur at the most once in a product appearing in the symmetric sums (38).

The Lagrangian parameters $\{\lambda_\nu, \mu\}$ are determined in principle by the constraints (30) if each one-electron state is identified by spectroscopic post-foil measurements. In practice this is hardly the case, the spectroscopic measurements leading only to the determination of the probabilities p_k in association with the identification of the configurations κ with the number of electrons, N_κ. Consequently, as a first approximation, the constraints (30a) and (30c) are needed. The corresponding probabilities p are obtained from Eq. (31) by putting all $\lambda_\nu = 0$. Hence $z = x_\nu = \exp(-\mu)$ in Eqs. (38) which results in the binomial distribution

$$P_k = \frac{(1+z)^M}{\vartheta}\binom{M}{k} p^k (1-p)^{M-k}, \tag{39}$$

with $p = z/(1+z)$. If N in Eqs. (38) is large in addition to M, then the distribution (39) can be shown to be very close to the Gaussian

$$P_k = \frac{1}{\sqrt{2\pi}\,\sigma} \exp\left[-\frac{1}{2}\left(\frac{k-\langle N\rangle}{\sigma}\right)^2\right], \tag{40}$$

where $\sigma = \sqrt{zM}\,/(1+z)$ and $\langle N\rangle$ is the mean number of electrons after the foil. The proof is based on the known relationship[34] of the Gaussian and the binomial distribution at large M and on the fact that since $\exp(-\mu)$ must be small $\vartheta \cong (1+z)^M$ and $\langle N\rangle = -(d\ln\vartheta/d\mu) \cong Mz/(1+z)$. The limiting form (40) is what one expects on general grounds since it can also be shown[25] that if a stochastic variable X can be reduced (i.e., the mean and variance are known), then the Gaussian $\exp(-\frac{1}{2}X^2)$ maximizes the information-theoretical entropy.

It has recently been found by Baudinet-Robinet *et al.*[35] that experimental charge-state distributions follow the Gaussian law extremely well for ions passing through carbon foils at energies larger than approximately 20 keV amu^{-1}. For energies less than 20 keV amu^{-1} and for gas targets a chi-squared distribution seems to account better for an enhancement of highly charged ions. From an information-theoretical point of view these results indicate that for high energies the ions are randomly excited to the extent that the atomic structure, represented by the second constraint (30b), does not influence the final charge distribution significantly. For lower energies and gas targets the skewness of the distribution may indicate an enhancement of the relative abundance of highly charged ions by autoionization. In addition, one expects a partitioning of the x_ν and consequently a grouping of the probabilities (38) into several Gaussians pertaining to the shell structure of the ions. But this is nothing but a chi-squared distribution with respect to the total number of electrons which is a sum of the number of electrons in each shell. Hence from a information-theoretical point of view the chi-squared distribution reflects the information about the atomic structure inherent in the constraint (30b) and in the autoionization process.

It follows also from Eqs. (38) that the probabilities P_k must fulfill some consistency relations. Using the notation

$$\langle x^n\rangle = \sum_{\nu=1}^{M} x_\nu^n \tag{41}$$

we may, for example, show by solving Eqs. (38) successively from $k = 1$ to 4 that

$$\begin{aligned}
\langle x\rangle &= y_1, \\
\langle x^2\rangle &= y_1^2 - 2y_2, \\
\langle x^3\rangle &= y_1^3 - 3y_1y_2 + 3y_3, \\
\langle x^4\rangle &= y_1^4 - 4y_2y_1^2 + 4y_3y_1 + 2y_2^2 - 4y_4,
\end{aligned} \tag{42}$$

where $y_k = P_k/P_0$ $(k = 1, 2, 3, 4)$. Evidently it may be checked under some circumstances whether the experimental ratios y_k make the sums on the right-hand side of Eqs. (42) positive. A failure indicates either inaccurate data (already corrected for autoionization) or an inadequacy of the one-electron representation in connection with the conjecture of random excitation. The procedure (42) can of course be continued to higher k.

Explicit considerations of the influence of the autoionization process on the final charge distributions require usually a division of the one-electron states into various classes within a given ion. Furthermore, one may introduce the central-field model explicitly for the identification of the one-electron states. We shall illustrate this procedure by considering the passage of He^+ ions through a foil. It is assumed that the alignment is not observed.

For helium, the following six classes can be formed, numbered consecutively from one to six: (1) He $1s^2$, (2) He $1snl$, (3) He $nln'l'$, (4) He^+ $1s$, (5) He^+ nl, and (6) He^{++}, where $n(n') \geq 2$. Out of these six classes (3) may decay into (4) or (5) by autoionization. Let us denote the total relative autoionization rate by δ. Since the Lagrangian parameters λ_ν do not depend on m_l we may also introduce the relative occupation probability $P_{nl} = \exp(-\lambda_{nl} - \mu)$. The charge distributions corres-

ponding to Eqs. (38), modified by the autoionization rate are then given by

$$P_0' = \vartheta^{-1},$$

$$P_1' = \vartheta^{-1}\left[2P_{1s} + R + \frac{\delta}{2}\left(R^2 - \sum_{n>1,l} g_{nl}P_{nl}^2\right)\right], \tag{43}$$

$$P_2' = \vartheta^{-1}\left[P_{1s}^2 + 2P_{1s}R + \frac{1-\delta}{2}\left(R^2 - \sum_{n>1,l} g_{nl}P_{nl}^2\right)\right],$$

where $g_{nl} = 2(2l+1)$ is the degeneracy and where

$$R = \sum_{n>1,l} g_{nl}P_{nl}.$$

The partition function ϑ is given by

$$\vartheta = (P_{1s} + R + 1)^2 - R - \tfrac{1}{2}\left(R^2 + \sum_{n>1,l} g_{nl}P_{nl}^2\right). \tag{44}$$

Equations (43) provide the link between the level populations and the final charge distribution in the case of random excitation. If the sum over P_{nl}^2 can be neglected in Eqs. (43), then P_{1s} and R are uniquely determined by the charge distribution. Otherwise Eqs. (43) can be used as a consistency test of measured level populations and charge distribution in helium and heliumlike ions for which $\delta \cong 1$. For that purpose the contribution from the level populations of high Rydberg states can be estimated using the n^{*-3} law (26).

IV. DISCUSSION

It has been shown that the maximum entropy principle predicts a level-population density matrix which is in accordance with a number of general features of the foil excitation including the n^* and T dependence of the excitation probabilities for high Rydberg states. The Gaussian or chi-squared character of the charge distribution also follows as well as its relation to level populations.

The evidence presented above indicates that in most cases studied the foil produces ions which can be considered randomly excited after the processes inside and at the back of the foil. This evidence is mainly concentrated to outer-shell excitations. However, the fact that the maximum-entropy density matrix both reproduces and organizes the experimental data does not provide a sufficient condition for random excitation. It just suggests a plausible alternative to detailed models of foil excitation with ill-defined range of validity. In a narrow sense Eqs. (22) would only represent a reparametrization of the observed intensities. The strength in the information-theoretical procedure lies in the fact that from a limited set of data it predicts a simple behavior of the Lagrangian parameters λ and μ which can be used to explain general features of the charge distribution and eventually of the radiation pattern in the unobserved or unresolved part of the ionic spectrum. With respect to the method it is useful to consider Planck's blackbody radiation law as an extreme example. As elucidated in Sec. II B the identification of the Lagrangian parameter with temperature makes it meaningless to invoke a detailed microscopic model of the interaction between the atoms in the wall and the radiation field. Consequently, inconsistencies and complexities in the behavior of the Lagrangian parameters would be an indication of rather distinct foil excitation mechanisms. Relations (42) for the charge distribution and corresponding ones for the level populations which follow from Eq. (31) are useful in this connection.

We recall that according to the maximum entropy principle a Rydberg series has, for large n, a level-population function

$$P_{nl}(T) = \mathrm{Tr}_{nl}\rho = 2(2l+1) \times \exp[-\lambda_0 - 3\ln n^* - \mu_l(T) - N_q\mu(T)]. \tag{45}$$

The notation is defined by Eq. (28). The general behavior, common to all series, as a function of T is determined by $\mu(T)$ whereas $\mu_l(T)$ accounts for differences between the various series. Independently of n the parameter $\mu_l(T)$ also determines how P_{nl} depends on l. Consequently, on a log-plot P_{nl} should be a universal function of l. The existing limited data[28,36] do not disagree with this conclusion.

However, it should be noted that Eq. (45) does not account for alignment. This property should be examined according to Eq. (14), i.e., any measurement which is made with a detector at a fixed angle θ with respect to the beam direction yields also information about

$$\mathrm{Tr}_{nl}\{\rho[3m^2 - l(l+1)]\}.$$

Considerations like those presented above can be made more systematic by using surprisal analysis.[14,15] This may be of use when more abundant and accurate data on the beam-foil excitation become available. The surprisal analysis is based on the use of Eq. (17) for a set, $\{\hat{T}_\mu\}$, of commuting operators. If $\{|\kappa\rangle\}$ is a common set of eigenstates we may write

$$\langle\kappa'|\hat{\rho}|\kappa\rangle = \delta_{\kappa',\kappa}\exp\left(-\sum_{\mu=0}^{M}\lambda_\mu T_\mu\right), \tag{46}$$

where T_μ are the corresponding eigenvalues. Suppose the Lagrangian parameters λ_μ are independent of κ over a range Γ of n_Γ values of κ. Then it follows from Eq. (46) that one can introduce the "surprisal"

$$-\ln\frac{P_\Gamma}{P_{0_\Gamma}} = -\ln\left(\frac{\mathrm{Tr}_\Gamma\rho}{\mathrm{Tr}_\Gamma\rho_0}\right) = \sum_{\mu=0}^{M}\lambda_\mu T_\mu , \tag{47}$$

where λ_0 is redefined in accordance with the definition of ρ_0, given by

$$\langle\kappa'|\hat{\rho}_0|\kappa\rangle = \delta_{\kappa'\kappa}\Big/\left(\sum_\Gamma n_\Gamma\right) . \tag{48}$$

Equation (47) has been extensively used by Levine and his co-workers[14,15] for the representation of experimental probabilities P_Γ in molecular collisions and heavy-ion-induced nuclear reactions. A remarkable fact is that often a single constraint is sufficient to account for the data in which case Eq. (47) is represented by a straight line in a plot of experimental $-\ln(P_\Gamma/P_{0_\Gamma})$ versus the dominating T_μ.

A surprisal corresponding to Eq. (45) is given for a fixed N_q by

$$-\ln\frac{P_{nl}(T)}{2(2l+1)n^{*-3}} = \lambda_0 + \mu_l(T) - \mu_{l_0}(T) , \tag{49}$$

where $\lambda_0 = -\ln[P_{n_0l_0}(T)/2(2l_0+1)n_0^{*-3}]$ defines a reference level (n_0l_0). In their analysis of the Kr VIII $4s\,^2S - 4p\,^2P$ decay curve and other measured decay curves Younger and Wiese[37] determined the relative population from an equation which corresponds to $\mu_l(T) = \mu_{l_0}(T)$ in Eq. (49). Hence one would interpret their choice as the one which the maximum entropy principle would give when the only constraint according to Eq. (27) is due to the normalization of the probabilities. The function $\mu_l(T) - \mu_{l_0}(T)$ arises according to Eq. (29) from the identification of the Rydberg series. It accounts for the anomalous features[28,36] in the l dependence. Under normal circumstances this is expected to be a simple universal function of l, for given T. As a contrast abrupt behavior of the surprisal indicates a specific mechanism, possibly a resonance.[29]

In conclusion, we suggest that beam-foil data should be analyzed using information theory along the lines which have been outlined in this paper. This may help to decide where the selective features in the foil excitation mechanism reveal themselves.

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