Interpretation of unresolved transition arrays in the soft-x-ray spectra of highly ionized molybdenum and palladium

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Previously published theoretical formulas for the mean wavelengths and spectral widths of unresolved transition arrays are applied to transitions of the types $3d^{N+1}$-$3d^N4p$ and $3d^{N+1}$-$3d^N4f$ observed in highly ionized spectra of Mo and Pd. Agreement with experiment is excellent and opens the way to the quantitative interpretation of the intensities.

I. INTRODUCTION

When heavy atoms are highly ionized in hot plasmas, the internal $3d$, $4d$, or $4f$ subshells become open. In the experimental spectra, the very numerous lines of transition arrays between configurations tend to merge together when the width of the array is small compared with its mean wavelength. This is the case of the $\Delta n \neq 0$ transitions, e.g., $3d^{N+1}$-$3d^N4f$.[1,2] Various broadening mechanisms (Doppler, Stark, etc.) contribute to the merging of the lines, but very little to the width of the transition array, originating in the splitting between the levels of the configurations.

Such arrays have been observed in a variety of spectra.[3–14] In a previous paper[1] (to be referred to hereafter as I) we have shown that these unresolved transition arrays (UTA) can be considered as statistical distributions of wave numbers (or wavelengths), with the "statistical weight" or "probability of occurrence" being here the line intensity. These statistical entities can be described by their different statistical moments. In I, we gave exact formulas for the first and second moments of the transition arrays l^{N+1}-l^Nl' and l^Nl'-l^Nl''. In this report, we wish to compare these results with two experimental spectra of molybdenum and palladium. Actually, the comparison with the Mo spectrum was already published in I, but the agreement with experiment was not quite satisfactory, due to an error in our spectrum plotting program.[15]

II. BRIEF SUMMARY OF THE THEORETICAL RESULTS

The moments μ_n of the distribution of wave numbers can be written as

$$\mu_n = \frac{\sum_{a,b} [\langle b \mid H \mid b \rangle - \langle a \mid H \mid a \rangle]^n \mid \langle a \mid Z \mid b \rangle \mid^2}{\sum_{a,b} \mid \langle a \mid Z \mid b \rangle \mid^2} ,$$

where a,b run over all the exact eigenstates of the Hamiltonian in the lower and upper configurations A and B, respectively, and Z is the z component of the dipole transition operator. The most useful quantities are the mean wavelength μ_1 and the variance $\sigma^2 = \mu_2 - (\mu_1)^2$, which is related to the spectral width of the UTA. In the development of μ_2, expressions like

$$Q = \sum_{a,b} \langle a \mid H \mid a \rangle \langle b \mid H \mid b \rangle \langle a \mid Z \mid b \rangle \langle b \mid Z \mid a \rangle$$

appear. We use the formalism of the second quantization[16] to evaluate them.

It was shown in I that the results for σ^2 can always be written as

$$\sigma^2 = \sum_i \mathcal{N}_i \left[\sum_{k,k'} x_i(k,k',l,l',\ldots) \times X_i^{kk'}(nl,n'l',\ldots) \right] , \quad (1)$$

where $\mathcal{N}_i$ are simple numerical factors depend-

ing on the number of equivalent electrons N, $x_i(k,k',l,l',\ldots)$ are combinations of $3j$, $6j$, and $9j$ symbols independent of N, and the X_i are products of Slater integrals of rank k and k'. There are as many values of i as there are products of Slater integrals (F^k, $G^{k'}$, etc.) for which the contribution does not vanish (see I). Spin-orbit integrals intervene in a similar manner. For the mean wavenumber of the arrays, it was found in I that

$$\mu_1 = E_{\rm av}(B) - E_{\rm av}(A) + \delta E \ ,$$

where $E_{\rm av}(B)$ and $E_{\rm av}(A)$ are, respectively, the average energies of the two configurations involved. δE is a shift originating in the weighting factor $|\langle a|Z|b\rangle|^2$, and is nonvanishing for the l^{N+1}-$l^N l'$ type. It is equal to a combination of Slater integrals and simple coefficients, and is proportional to N. For Mo XVI ($3d^9$-$3d^8 4f$), it was found to correspond to 0.8 Å (at $\lambda \simeq 31$ Å), and for Cs XI ($4d^9$-$4d^8 4f$) its value is 28 Å (at $\lambda \simeq 105$ Å).[17]

All these results are valid whatever the physical coupling scheme prevailing in the configurations A and B.

III. COMPARISON WITH EXPERIMENT

Figure 1 shows a spectrum of Mo, obtained from a high-power vacuum spark,[6] compared to the theoretical UTA. In Fig. 2, the experimental spectrum of Pd was obtained in a laser-produced plasma with a somewhat lower spectral resolution. In these two spectra, many ionization stages may be seen. These are present at different times and places of the plasmas. For each ionization stage, the ground configuration $3d^{N+1}$ gives rise to transitions of the types $3d^{N+1}$-$3d^N 4p$, $3d^{N+1}$-$3d^N 4f$, etc. The formulas developed so far give only the first and second moments of the UTA. In order to compare to experiment and to draw curves, we have to assume a profile, which we choose as Gaussian for the sake of simplicity. The necessary integrals and averages of configurations have been evaluated *ab initio* by the relativistic parametric-potential method.[18,19] In order to use our nonrelativistic formulas, the relativistic Slater integrals were averaged following the prescription of Larkins.[20] For these high stages of ionization, a nonrelativistic model would not give as good an agreement with experiment.[21] The aim of this paper being the comparison with experimental mean wavelengths and spectral widths, and not with the intensities of the UTA, we took the latter as free parameters to obtain the best fit.

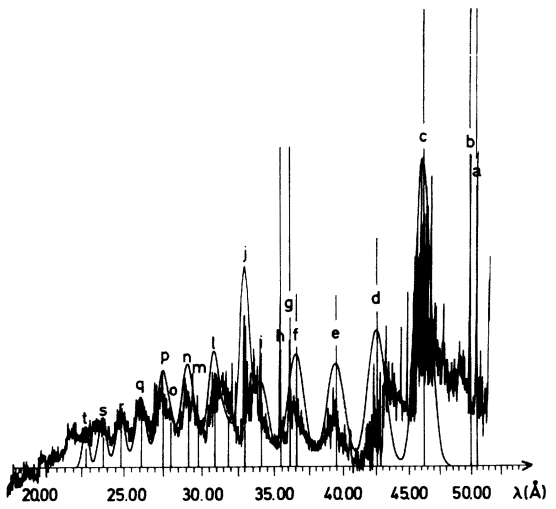

FIG. 1. Experimental spectrum of Mo in spark (Ref. 6) compared with theoretical UTA. The intensity of each array was adjusted to obtain the best fit. $3d^{N+1}$-$3d^N 4p$ transitions: a,b—Mo XV; c,d,e,f,i,k,m,o—Mo XVI to Mo XXIII. $3d^{N+1}$-$3d^N 4f$ transitions: g,h—Mo XV; j,l,n,p,q,r,s,t—Mo XVI to Mo XXIII.

The agreement for Mo is satisfactory, and it is even better for Pd. This means that we are able to extract from the experimental spectrum the ratio of intensities of the $3d^{N+1}$-$3d^N 4p$ transitions to $3d^{N+1}$-$3d^N 4f$ (with adequate corrections for the efficiency of the spectrograph and the detectors, etc.). This ratio generally depends on the plasma parameters T_e (electronic temperature) and n_e (electron density). In the framework of an appropriate model for level populations in the plasma, one could then estimate n_e and T_e, thus obtaining a plasma diagnosis.

Coming back to the spectra, we note some small but interesting discrepancies. In Fig. 1, around 31 Å, the experimental array seems shifted toward long wavelengths. In Fig. 2, an analogous discrepancy appears at 24 Å. Both lie near the Co-type ionization stages (Mo XVI and Pd XX, respectively). Another feature common to the two spectra is the appearance of a very broad "continuum" in the vicinity of the Fe-like $3d^{N+1}$-$3d^N 4p$ transitions, extending beyond the Ni-like array. These features probably originate in some satellite arrays involving doubly excited configurations. It is clear, however, that without a satisfactory model for the simple $3d^{N+1}$-$3d^N 4p$ and $3d^{N+1}$-$3d^N 4f$ transitions we would not have been able even to point out the problematic features.

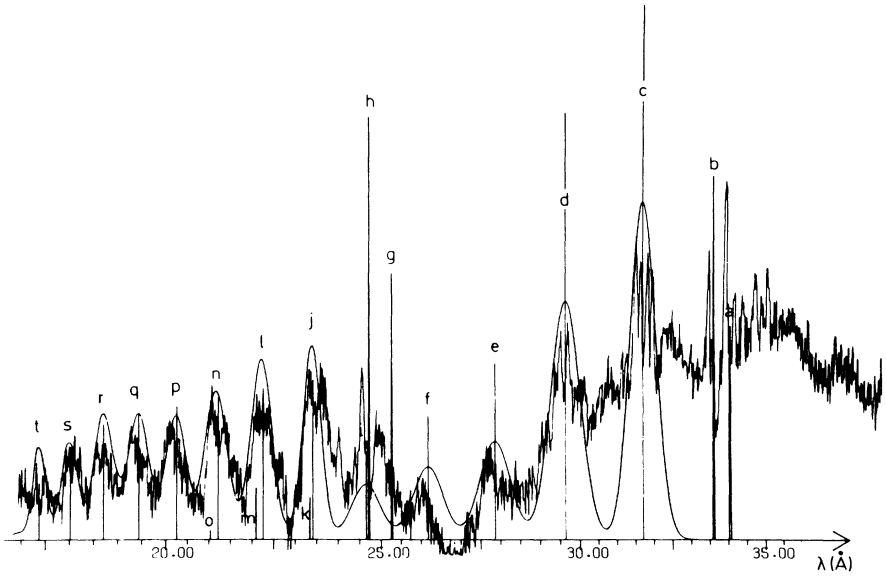

FIG. 2. Experimental spectrum of Pd in laser-produced plasma (this work), compared with theoretical UTA. The intensity of each array was adjusted to obtain the best fit. $3d^{N+1}$-$3d^N4p$ transitions: a,b—Pd XIX; c,d,e,f,i,k,m,o—Pd XX to Pd XXVII. $3d^{N+1}$-$3d^N4f$ transitions: g,h—Pd XIX; j,l,n,p,q,r,s,t—Pd XX to Pd XXVII.

IV. CONCLUSION

We have presented here examples of application of a new model, proposed in I, describing unresolved transition arrays (UTA) as statistical distributions of wave numbers. The comparisons with the experimental spectra of Mo and Pd (Figs. 1 and 2) show that this model is adequate for interpreting quantitatively the spectra, pointing out missing transition arrays (in the computed spectrum), and enabling the extraction of experimental intensity ratios of UTA. Thus, our statistical model opens the possibility for performing spectroscopical plasma diagnosis *without identification of single lines.* This could obviously have far-reaching consequences on the design of spectrometers for hot-plasma diagnosis.

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[15]Please note also a typing error in the formula D_7 of Table I in I: the $6j$ symbol should read

$$\begin{Bmatrix} l' & l' & k \\ l & l & k' \end{Bmatrix}.$$

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