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Measurements of line overlap for resonant photopumping of $4 \rightarrow 4$ transitions in highly charged nickel-like ions

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The wavelength of the $6f_{7/2} \rightarrow 3d_{5/2}$ transition in several nickel-like ions has been measured on the Livermore electron-beam ion trap with a precision as high as 27 ppm in order to assess their utility in a resonant photopumping scheme to achieve x-ray lasing below 80 Å. An interpolation of the results is presented, which provides an estimate of the transition energies for ions between Dy ($Z=66$) and U ($Z=92$). The closest line coincidence was found for the $3d$-$6f$ transition in nickel-like Hf, which differs by 1.2 mÅ from the wavelength of the Ly-α_2 transition in hydrogenic Cl, and for the $3d$-$6f$ transition in Tl, which differs by 0.8 mÅ from Ly-α_1 in hydrogenic Ca.

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I. INTRODUCTION

The development of soft-x-ray lasers has been of great interest in recent years. Amplification and lasing has been demonstrated in arrangements utilizing collisionally pumped neonlike or nickel-like ions as well as in recombination-based schemes involving hydrogenic or lithiumlike ions [1–4]. Resonant photopumping has been proposed as an alternative method to achieve lasing in the x-ray regime [5,6], and there is evidence that resonant photopumping plays an important role in the neonlike titanium laser at 326.5 Å [7].

Most proposed photopumping schemes are based on chance coincidences between transitions in the lasant ion and the pump. An exception is the photopumping scheme proposed by Nilsen for achieving gain in highly charged nickel-like ions [8]. It consists of a family of resonant pairs working off the ground state of the lasant ion. In particular, it is proposed to pump the $(3d_{5/2}^{-1}6f_{7/2})_{J=1}$ level in certain highly charged nickel-like ions by the Ly-α transitions from hydrogenic ions with atomic number Z between 17 and 23. Here $3d_{5/2}^{-1}$ denotes a vacancy in the filled $3d^{10}$ shell. In this scheme, decay of the $(3d_{5/2}^{-1}6f_{7/2})_{J=1}$ level to the $(3d_{5/2}^{-1}4d_{5/2})_{J=0}$ and $(3d_{5/2}^{-1}4d_{5/2})_{J=1}$ levels then enables lasing between the $4d_{5/2}$ and $4p_{3/2}$ levels. The wavelengths of the resultant lasing transitions range from 23 to 80 Å [8].

The effectiveness of a photopumping scheme depends on the degree of overlap between the pump and absorber transition. Thus, to assess the scheme proposed in Ref. [8] accurate experimental data are needed on the transition energies of the $6f_{7/2} \rightarrow 3d_{5/2}$ in various nickel-like ions. The M-shell x-ray spectrum from nickel-like ions has been measured using a variety of laboratory sources such as tokamaks, laser-produced plasmas, and accelerators [9-13]. Identification of $6 \rightarrow 3$ transitions were made by Tragin *et al.* for nickel-like Ta, W, Re, Pt, Au, Hg, and Pb excited by laser irradiation, and transition energies were measured with an accuracy between 400 and 700 ppm [14]. Here we report measurements of the $6f_{7/2} \rightarrow 3d_{5/2}$ transitions from nickel-like Tm, Hf, Re, Pb, and Th recorded on the Livermore electron-beam ion trap (EBIT). The accuracy of the measurements is between 27 and 45 ppm.

II. EXPERIMENT

The Livermore EBIT uses an electron beam to generate, trap, and excite highly charged ions in a 2-cm-long trapping region, as described in detail by Levine and Marrs [15,16]. Thus, the method of x-ray generation in EBIT is the inverse of the beam-foil effect employed on heavy-ion accelerators. The energy of the electron beam can be varied between 500 and 40000 eV to select the ionization state of interest. Metallic elements are injected into the trap with a metal vapor arc described by Brown *et al.* [17]; gases are introduced by ballistic injection.

X rays generated in electron-ion interactions are analyzed using EBIT's von Hámos-type crystal spectrometer described by Beiersdorfer *et al.* [18]. With this instrument we determine the wavelength of a particular transition relative to a set of reference lines, e.g., Refs. [19,20].

We have performed separate experiments to measure the line overlap between four pairs of ions: nickel-like Tm and hydrogenic S, nickel-like Hf and hydrogenic Cl, nickel-like Re and hydrogenic Ar, as well as nickel-like Th and hydrogenic V. In each case we measured several spectra, alternating Ly-α spectra with nickel-like spectra to ascertain reproducibility. In a fifth experiment we studied the $6f \rightarrow 3d$ transition in nickel-like Pb. It falls in a region that contains the Ly-γ and Ly-δ transitions of hydrogenic Ar, and we use these lines as references to determine its wavelength. The type of crystal employed in each experiment and its radius of curvature are listed in Table I. Recording a spectrum typically required 20 min. Like argon and chlorine, sulfur was introduced into EBIT with the ballistic gas injection system by exploiting its high vapor pressure at room temperature. Representative spectra of nickel-like Re and hydrogenlike Ar are shown in Fig. 1. Ions in EBIT are virtually at rest, with a temperature that is predicted by modeling codes to be less than a few hundred electron volts [21]. Thus our measurements are unaffected by Doppler shifts and the line shape is mostly instrumental.

TABLE I. Measured wavelengths of $6f_{7/2} \rightarrow 3d_{5/2}$ transitions in various nickel-like ions and comparison to previous measurements. The uncertainty in the last digit is given in parentheses. R is the radius of curvature of the crystal employed in the von Hámos spectrometer.

Element	λ^a (Å)	λ^b (Å)	Spectrometer specifications	Hydrogenic reference lines (Å)
Tm	4.740 13(14)		Si(111), $R=100$ cm	S^{15+}: 4.727 35[c], 4.732 76[c]
Hf	4.191 88(11)		Si(111), $R=100$ cm	Cl^{16+}: 4.185 33[c], 4.190 74[c]
Re	3.734 16(17)	3.734(2)	Si(111), $R=100$ cm	Ar^{17+}: 3.731 10[c], 3.736 52[c]
Pb	2.920 48(10)	2.920(2)	LiF(200), $R=75$ cm	Ar^{17+}: 2.987 32[d], 2.987 76[d]
				2.917 49[e], 2.917 56[e]
Th	2.279 31(8)		LiF(200), $R=75$ cm	V^{22+}: 2.277 60[c], 2.283 02[c]

[a]Present measurement.
[b]N. Tragin *et al.*, Ref. [14].
[c]Values for Ly-α transitions from Johnson and Soff, Ref. [22].
[d]Values for Ly-γ transitions from Garcia and Mack, Ref. [23].
[e]Values for Ly-δ transitions from Garcia and Mack, Ref. [23].

The wavelength λ of the nickel-like transition recorded in channel N is determined from the relation

$$\lambda = 2d \sin[\theta_0 + k(N - N_0)] . \quad (1)$$

Here d is the lattice spacing of the crystal, k is the dispersion determined from the separation of the Lyman lines, and θ_0 is the Bragg angle of the Lyman line situated in channel number N_0. The wavelengths of the Ly-α lines were set to values calculated by Johnson and Soff [22]; those of Ly-γ, Ly-δ, and Ly-ϵ were set to values calculated by Garcia and Mack [23]. The results are listed in Table I. Because the hydrogenic reference lines are very close, uncertainties in the dispersion do not affect the accuracy with which we can measure the nickel-like transitions. Instead, the experimental certainty is mainly limited by the accuracy with which we can determine the center position of a given line. For the rhenium line, shown in Fig. 1, this value is about 0.1 mÅ, which is about a tenth of the linewidth. Accounting for similar uncertainties in the position of the hydrogenic reference lines, the overall error is estimated as 0.17 mÅ or 45 ppm. Yet higher accuracy is obtained in the measurements of the other nickel-like transitions—37 ppm for Th, 35 ppm for Pb, 31 ppm for Tm, and 27 ppm for Hf.

In the case of Re and Pb we can compare our result to that of an earlier measurement by Tragin *et al.* [14] obtained from a laser-irradiated plasma source (cf. Table I). The measurements agree within error limits.

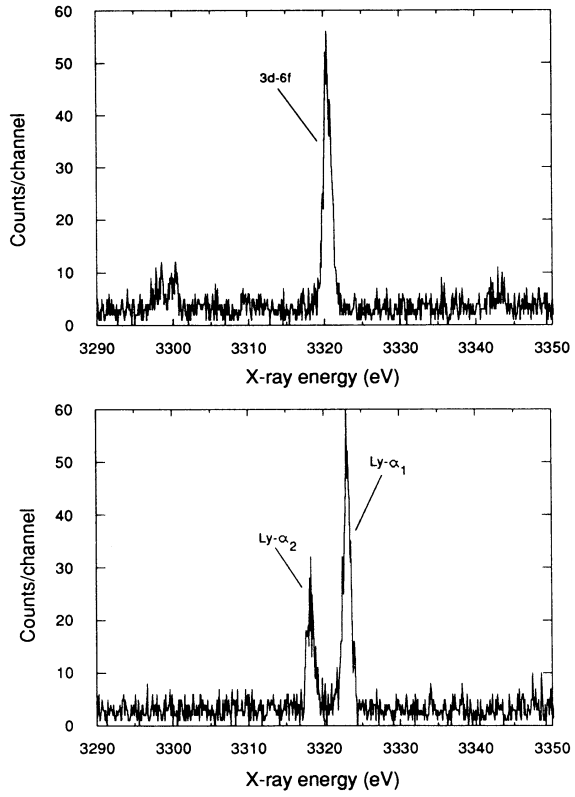

FIG. 1. Typical spectra observed with the high-resolution von Hámos spectrometer. Top: $6f_{7/2} \rightarrow 3d_{5/2}$ transition in nickel-like Re^{47+}. Bottom: $2p \rightarrow 1s$ transitions in hydrogenlike argon. A Si(111) crystal with a 100-cm radius of curvature was used in the measurement.

III. DISCUSSION

In first approximation, the energies of the Ly-α transitions scale with atomic number Z as $0.75Z^2 I_H$, where I_H is the Rydberg energy. Similarly, the energies of the $6f_{7/2} \rightarrow 3d_{5/2}$ transition scale as $0.75(Z^*)^2 I_H$ in the high-Z limit, where $Z^* = (Z-21)/3$, as shown by Nilsen [8]. Deviations from this scaling arise due to relativistic effects, configuration interaction, etc., and are smaller than 1%.

In Fig. 2 we have plotted the percent deviation of the Ly-α and $6f \rightarrow 3d$ transitions from the $0.75(Z^*)^2 I_H$ scaling. We fitted the deviation of the measured nickel-like transitions to a third-order polynomial in Z^*, which we use to interpolate among the wavelengths of the $6f \rightarrow 3d$ transitions. A cubic is chosen because it represents the lowest-order polynomial that fits all data points; moreover, higher-order polynomials yield a fit virtually identical to that of the cubic. The results of the interpolation

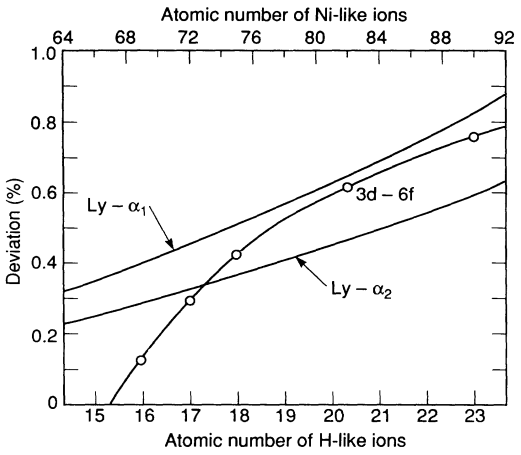

FIG. 2 Percent deviation of the Ly-α and $6f_{7/2} \rightarrow 3d_{5/2}$ lines from a $0.75(Z^*)^2 I_H$ scaling as a function of atomic number Z, where $Z^* = Z$ for the hydrogenic transitions and $Z^* = (Z - 21)/3$ for the nickel-like transitions. The measured values for the nickel-like transitions are indicated by open circles and are fitted by a cubic polynomial to allow determination of unmeasured wavelengths.

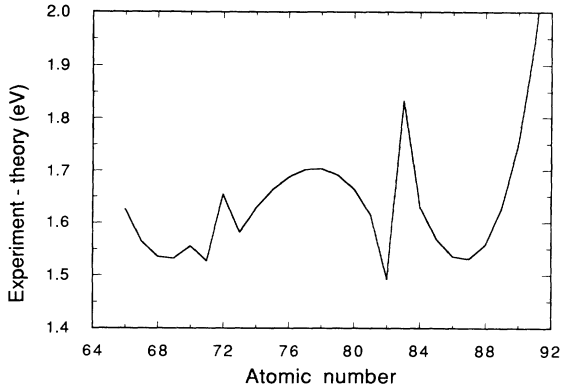

FIG. 3. Differences between the $6f_{7/2} \rightarrow 3d_{5/2}$ transition energies obtained from the cubic interpolation of the experimental data in Fig. 2 and values obtained with a multiconfigurational Dirac-Fock calculation.

are listed in Table II for nickel-like ions from Dy ($Z = 66$) to U ($Z = 92$). For comparison, we also list the results of a multiconfiguration Dirac-Fock calculation using the code of Grant *et al.* [24]. The calculations were done in the extended-average-level approximation using the 36 relativistic orbitals up to $6h_{11/2}$ and the 427 relativistic configurations which include the ground state and all the $n = 4, 5, 6$ singly excited states.

The differences between the calculated and fitted values are plotted in Fig. 3. On average, the calculated energies are about 1.62 eV smaller than those from the cubic fit. The differences do not vary smoothly with atomic number, however, but "discontinuities" as large as 0.3 eV disrupt a smooth variation. Since the polynomial fit to the data is a smooth function, any discontinuity must stem from nonsystematic effects in the theory. A possible nonsystematic effect is given by avoided level crossings. The calculations predict that the $(3p_{3/2}^{-1} 5d_{5/2})_{J=1}$ level crosses the $(3d_{5/2}^{-1} 6f_{7/2})_{J=1}$ level at $Z = 72$, while the $(3d_{3/2}^{-1} 6p_{3/2})_{J=1}$ level crosses the $(3d_{5/2}^{-1} 6f_{7/2})_{J=1}$ level at $Z = 83$. The location of these avoided crossings indeed corresponds to the location of the discontinuities in Fig. 3.

Photopumping works best for perfect line overlap, with opacity and Doppler broadening usually determining the allowable mismatch. Because of the definition of Z^* only every third nickel-like ion along the isoelectronic sequence can possibly fulfill the resonance condition for photopumping. From Fig. 2 we see that the nickel-like transitions of elements with atomic number greater than $Z = 72$ fall in between the Ly-α transitions; those below $Z = 72$ have energies less than those of the hydrogenic lines. The figure shows that the best overlap occurs for the combinations hydrogenlike Cl and nickel-like Hf ($Z = 72$) and hydrogenlike Ca and nickel-like Tl ($Z = 81$). The separation of the $6f_{7/2} \rightarrow 3d_{5/2}$ transition of nickel-like Hf from Ly-α_2 is a mere 0.81 eV (1.2 mÅ); the separation of the nickel-like Tl transition from Ly-α_1 is only 1.07 eV (0.8 mÅ). This compares to a Doppler-broadened linewidth of 0.86 eV (1.2 mÅ) for hydrogenlike Cl ions with a temperature of 500 eV and of 1.6 eV (1.2 mÅ) for hydrogenlike Ca ions with a temperature of 1 keV. If the appropriate plasma conditions were attained, gain and lasing would result at 70.8 Å [$(3d_{5/2}^{-1} 4d_{5/2})_{J=0} \rightarrow (3d_{5/2}^{-1} 4p_{3/2})_{J=1}$ transition] and 79.8 Å [$(3d_{5/2}^{-1} 4d_{5/2})_{J=1} \rightarrow (3d_{5/2}^{-1} 4p_{3/2})_{J=1}$ transition] in the Hf-Cl combination and at 54.3 and 62.2 Å, respectively, in the Tl-Ca combination.

TABLE II. Energies of $6f_{7/2} \rightarrow 3d_{5/2}$ transitions in nickel-like ions. The fitted values are obtained from the interpolation of the experimental data in Fig. 2. The MCDF values are from a multiconfiguration Dirac-Fock calculation.

Atomic number	Fit (eV)	MCDF (eV)	Atomic number	Fit (eV)	MCDF (eV)
66	2294.10[a]	2292.47	80	3969.76	3968.10
67	2398.98[a]	2397.41	81	4106.46	4104.84
68	2506.15[a]	2504.62	82	4245.42	4243.92
69	2615.62	2614.09	83	4386.65	4384.82
70	2727.37	2725.82	84	4530.16	4528.53
71	2841.41	2839.88	85	4675.96	4674.39
72	2957.72	2956.06	86	4824.05	4822.51
73	3076.30	3074.71	87	4974.44	4972.91
74	3197.14	3195.52	88	5127.14	5125.58
75	3320.26	3318.59	89	5282.17	5280.54
76	3445.63	3443.95	90	5439.53	5437.78
77	3573.27	3571.57	91	5599.25[a]	5597.31
78	3703.17	3701.47	92	5761.34[a]	5759.12
79	3835.34	3833.65			

[a]Extrapolated values.

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