J. Phys. B: At. Mol. Opt. Phys. **40** (2007) 167–176 doi:10.1088/0953-4075/40/1/015

Radiative parameters for some transitions arising from the $3d^94d$ and $3d^84s^2$ electronic configurations in Cu II spectrum

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Received 12 October 2006, in final form 6 November 2006
Published 20 December 2006
Online at stacks.iop.org/JPhysB/40/167

Abstract

Transition probabilities of 41 transitions originating from the $3d^94d$ and $3d^84s^2$ electronic configurations of singly ionized copper have been determined using laser-induced breakdown spectroscopy. The Cu II ions have been produced by laser ablation. The experimental relative transition probabilities have been converted into an absolute scale using measured branching fractions and theoretical radiative lifetimes of the corresponding upper states obtained by a relativistic Hartree–Fock method taking core-polarization and configuration interaction effects into account. Comparison of the new results with previously available data is also presented.

1. Introduction

In astrophysics, singly ionized copper (Cu II) has been observed in different objects. The Cu abundance in interstellar H I clouds has been derived from Cu I and Cu II transitions [1]. For a long time, the Cu II spectrum has been observed in different types of stars [2] e.g. in Ap stars [3] or in Bp stars of the Hg–Mn subgroup [4, 5].

Although many works have been devoted to the investigation of trends of Cu and Zn abundances in low-metallicity stars, the nucleosynthesis mechanisms controlling the production of these elements is still somewhat unclear and puzzling [6]. Observational data provided by Gratton and Sneden [7] and by Sneden and Crocker [8] favoured a primary-like process of Cu production (mechanisms yielding constant enrichment relative to iron) and a secondary-like process (iron seeds from previous stellar generations and enrichment proportional to the iron content). More recently, additional abundances for Cu in halo and disk stars were provided by different groups e.g. by Sneden *et al* [9], by Blake *et al* [10], Cunha *et al* [11] and by Hill *et al* [12]. Sneden *et al* [9] suggested that the chemical evolution of Cu and Zn might be ascribed mainly to the weak s-process (slow capture of neutrons in nucleosynthesis). This conclusion was questioned and evidence for a large contribution from relatively long-lived processes (type Ia supernovae) was provided by Raiteri *et al* [13] and Matteucci *et al* [14]. Production of these elements in significant amounts by the major nuclear burning processes in massive stars was also advocated by Timmes *et al* [15]. It has been inferred recently [6], from a study of a very large sample (90) of metal-poor stars, that the production of Cu and Zn requires a number of different sources like neutron capture in massive stars, s-processing in low and intermediate mass stars and explosive nucleosynthesis in various supernova types.

It is well known that a quantitative analysis of stellar spectra and, more particularly, a detailed abundance determination in stellar atmospheres relies heavily on radiative parameters and, particularly, on transition probabilities. With the present set of data, we aim at providing new quantitative information on Cu II spectrum in order to establish more reliable constraints on the still open problem involved in Cu and Zn formation.

Radiative data for Cu II are also needed in other fields of physics. In plasma physics, the determination of transition probabilities for Cu II lines is justified by the extensive use of copper as an electrode material. Shapes and intensities of lines emitted by copper sputtered or evaporated from the electrodes are frequently considered for plasma diagnostics [16]. Despite this need, a relatively small number of papers has been devoted to the investigation of transition probabilities in Cu II, particularly for the transitions considered in the present paper (see further).

For atomic structure calculations, Cu II is an interesting medium-heavy element for which a strong interplay between the relativistic and correlation effects is to be expected and, consequently, these effects cannot be treated independently. Over the years, a number of theoretical studies have been devoted to the investigation of the spectrum of Cu II (see hereafter). Most of the earlier computations however have been performed either in a nonrelativistic scheme or by making use of semi-empirical approaches. In addition, most of the experimental and theoretical data about the radiative lifetimes and transition probabilities of the excited states of Cu II were devoted to states belonging to low lying $3d^94s$, $3d^94p$ and $3d^95s$ electronic configurations.

For these reasons, we report in the present paper on the determination of transition probabilities for 41 transitions originating, in emission, from the $3d^94d$ and $3d^84s^2$ electronic configurations little considered up to now in the scientific literature.

2. Previous works

Singly ionized copper belongs to the nickel isoelectronic sequence. It has two stable isotopes (^{63}Cu and ^{65}Cu) and its ground state is $[Ar]3d^{10}\,{}^1S_0$. The low-lying electronic configurations are $3d^{10}$, $3d^94s$, $3d^94p$, $3d^84s^2$, $3d^95s$, $3d^84s4p$ and $3d^94d$ (figure 1). Experimentally, the excitation energies of these configurations have been known for a rather long time [17, 18]. These levels have been derived from different spectral analyses due to Shenstone [19], Reader *et al* [20], Carter [21], Kaufman and Ward [22] and Ross [23] covering the range extending from the far UV up to the infrared region. Above 120 000 cm^{-1}, the experimentally known energy levels extend the $3d^9nl$ Rydberg series up to 10s, 7p, 8d, 7f and 7g, respectively [17, 18].

On the experimental side, the first transition probabilities in the Cu II spectrum (nine lines) were published by Corliss and Bozman [24] who determined *A*-values from arc measurements for spectral lines of 70 elements, including copper. Kock and Richter [25] measured transition

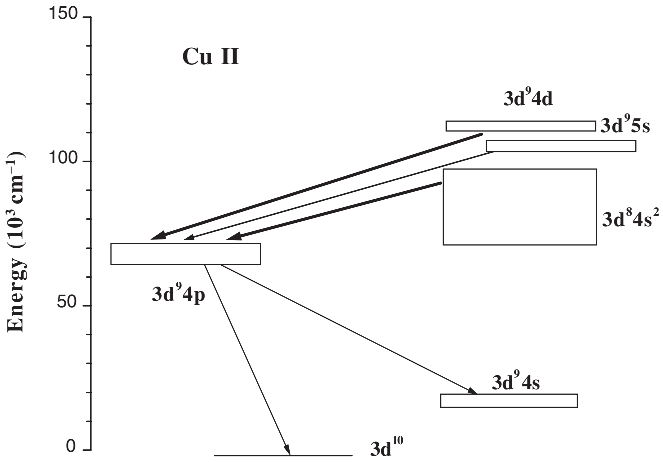

Figure 1. Partial energy level diagram of Cu II. The transitions considered in the present work are shown by arrows.

probabilities of some Cu II 4p–5s and 4s–4p transitions using a wall-stabilized electric arc under LTE conditions. A multichannel delayed coincidence method with crossed atomic and pulsed electron beams has been used in [26] to measure radiative lifetimes of the excited 5s $^3D_{2,3}$ states. The beam-foil method has allowed the determination of the radiative lifetimes of some states belonging to 4p, 4f and 5s configurations [27, 28]. The transition probabilities for transitions between the ground state and the low lying $3d^94s$ $^1D_2$, $^3D_2$ metastable levels of Cu II and the radiative lifetimes of these excited levels were experimentally investigated by Prior [29] with an electrostatic ion trap. The radiative lifetimes of excited states belonging to the configurations $3d^94p$ and $3d^95s$ have been measured by Kono and Hattori [30] with the delayed-coincidence method. Transition probabilities for 4s–4p and 4p–5s transitions were obtained from branching fraction (BF) measurements with a hollow-cathode lamp. Relative transition probabilities of $3d^94p$–$3d^84s^2$ spectral lines were deduced by Neger and Jäger [31] in the visible spectral region using an axial discharge with an exploding copper wire. The data were normalized with the transition at 455.592 nm. Transition probabilities for spectral lines of Cu II in the spectral region 200.0–300.0 nm, were determined in [32] from measurement of BFs for 4s–4p and 4p–5s transitions, using a high-frequency hollow electrode discharge. Experimental lifetimes of levels belonging to the $3d^94p$ configuration of Cu II were obtained by Pinnington *et al* [33] with the beam-laser method and were compared to relativistic Hartree–Fock calculations. The radiative lifetimes of 5p $^3P^\circ_1$, 4p $^1D^\circ_2$ and 5f $^1F^\circ_3$ levels of Cu II have been investigated also with the beam-gas technique [34].

On the theoretical side, transition probabilities for some forbidden lines were obtained by Garstang [35] more than 40 years ago. The oscillator strength of the resonant $3d^{10}$ ^1S–$3d^94p$ $^1P^\circ$ transition ($\lambda = 135.8$ nm) of Cu II was investigated in [36] with the MCHF method. The transition probability for the $3d^{10}$ ^1S–$3d^94s$ ^1D E2 transition was evaluated with a many-electron theory by Beck [37]. The *A*-values of the 4s–4p transitions and the radiative lifetimes of the $3d^9nl$ ^3L (l = s, p, d, f, g) levels published by Theodosiou [38] were computed with a relativistic potential to describe the ionic core. The calculations of the radiative lifetimes for levels belonging to the $3d^94p$ configuration were performed by Loginov [39] in intermediate coupling with a Hartree–Fock method. Calculations of oscillator strengths of an extensive set of transitions, including those arising from $3d^84p^2$,

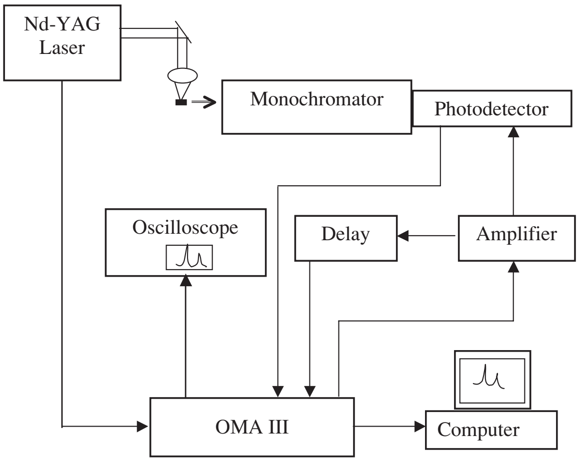

Figure 2. Experimental set-up used for branching fraction determination. For a description, see the text.

$3d^84s5s$, $3d^84s4p$ and $3d^84s4d$ configurations, were published by Donnelly *et al* [40]. The $3d^{10}$–$3d^94p$ resonance and intercombination transitions of Cu II were studied by using large-scale multiconfiguration Dirac–Fock calculations [41]. Relativistic many-body perturbation theory was applied for calculating the oscillator strengths of Ni-like ions (including Cu II) and the dependence of oscillator strengths for transitions between the ground state and 4p, 4f, 4s, 4d and 5f configurations as functions of the nuclear charge Z was investigated by Safronova *et al* [42]. The *f*-values of the transitions emitted from $3d^94p$, 5p, 5s, 4d, $3d^84s4p$ and $3d^84s^2$ configurations and theoretical lifetimes of the $3d^94p$ configuration have also been evaluated with the HFR approach including core-polarization effects [43].

In the literature there are only two theoretical papers [43, 44] devoted to the determination of radiative parameters of the $3d^84s^2$ levels in Cu II. In the first one [43], the HFR approach was applied and, in the second one [44], transition probabilities of two-electron $3d^94p$–$3d^84s^2$ transitions in Cu II spectrum were investigated with a Hartree–Fock method.

The transitions emitted from the 4d electronic configuration of Cu II have been studied in four publications [38, 40, 43, 45] in which different theoretical approaches were considered i.e. the HFR method [43], the CIV3 code [40] and a numerical Coulomb approximation [38, 45].

The main purpose of the present paper is to provide an experimental basis to the transition probabilities of $3d^94p$–$3d^94d$ and $3d^94p$–$3d^84s^2$ transitions and to compare these new and improved results with those published previously.

3. Experiment

In the present work, transition probabilities in Cu II have been obtained experimentally using laser-induced breakdown spectroscopy (LIBS). The experimental set-up is shown in figure 2.

The source of free copper atoms and ions in different stages of ionization is a laser-induced plasma. A Nd:YAG laser (YG 585 Quantel) working in Q-modulation regime was used for that purpose. The laser generated 160 mJ pulses of 7 ns duration at 20 Hz and 1064 nm wavelength. The laser beam was focused at right angle onto the Cu target. The experiment was performed in a vacuum chamber in a controlled argon atmosphere (∼8 Torr), which limits the plasma

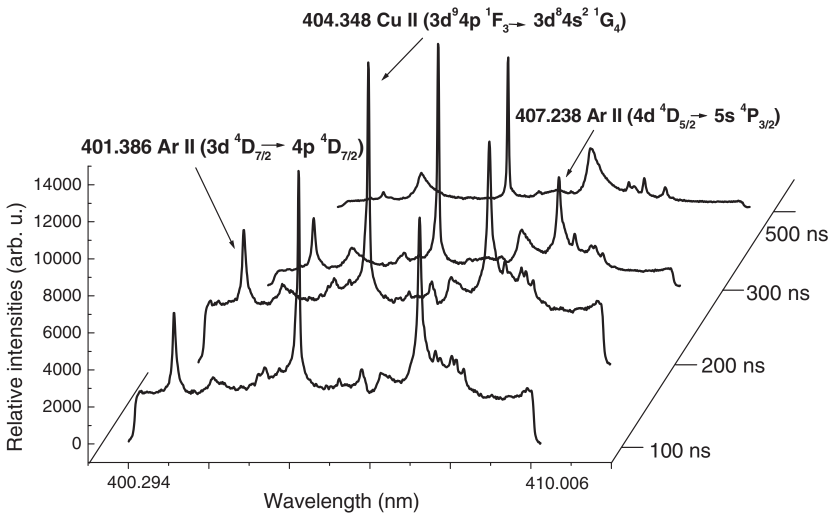

Figure 3. Time evolution of the copper spectrum in the region 400 to 410 nm for delay times ranging from 100 ns up to 500 ns.

cloud in the space. The target was placed on a stand which can be shifted in the horizontal and vertical directions so that the laser beam was focused precisely on its surface. After the registration of each spectrum, the target was rotated in order to avoid a hit of the laser beam on the same surface position. This could influence the intensity of the spectral lines and could lead to the destruction of the sample as well. The light arising from the plasma region was sent onto the entrance slit of a 1 m grating Czerny-Turner monochromator with 2400 grooves mm^{-1} and 0.03 nm resolution. A time-resolved optical multichannel analyser (OMA III, EG&G) with 1024 channels was used for the recording of the investigated spectra. A spectral region of about 10 nm in different delay times after the laser pulse (figure 3) was registered during each record. The electronic trigger and the laser Q-switch were synchronized.

The spectral response of the system in wavelength ranges 200–400 nm and 350–600 nm was determined using standard deuterium and tungsten lamps, respectively. This calibration was also checked by measuring the BFs of well-known Ar I and Ar II spectral lines. The response of the OMA photodiode array was also determined. The mode of calibration of the system was described previously [46] and will not be repeated here. The total error due to the calibration is estimated to about 6%.

To obtain the best signal-to-noise ratio, each spectrum was made at 100, 200, 300 and 500 ns after the laser pulse. We adopted the spectra recorded with a delay of 200 ns for the measurement of the transition probabilities because of its best signal-to-noise ratio.

The main broadening mechanism in the plasma is the Stark broadening due to charged perturbers that affects the Lorentzian profiles; it is one order of magnitude larger than the Doppler mechanism due to the thermal motion of the emitting atoms or ions which affects the Gaussian profiles. The convolution of both profiles was adopted to fit the observations. As the mechanisms act independently, the convolution was the best option considering an intensity derived from the contribution of the atoms in a Lorentzian distribution $L(x)$, but centred on a different frequency because of the Doppler shift $G(x - y)$. Accordingly, the wings depend on $L(x)$ while the peak and the full width at half maximum (FWHM) depend on $G(x - y)$. For our experimental set-up, we were able to fit lines down to FWHM $\sim$0.32 Å.

The investigated spectral lines were analysed by a software which allows us to fit the profiles of the spectral lines to Voigt profiles and to determine their characteristics. It was possible to separate closed or overlapping spectral lines. After subtraction of the background and treatment of the investigated spectrum, the relative intensities of the spectral lines were obtained with an adequate computer programme. The values from eight different measurements were averaged to determine the final intensity of each line.

The investigated sample was made from an alloy of copper and zinc to avoid self-absorption effects. In the present experiment, the possibility of overlap of the investigated spectral lines with Ar I, II or other copper lines, was considered. The experiment was carried out with a glass filter in front of the entrance slit of the monochromator to avoid the possible influence of second-order transitions where necessary.

4. HFR calculations

Theoretical estimates of the radiative lifetimes of levels belonging to $3d^94d$ and $3d^84s^2$ configurations and of BFs of the depopulating transitions were obtained with a pseudo-relativistic Hartree–Fock (HFR) model [47]. The computational procedure used in the present work for Cu II was exactly the same as that described in our previous paper [43] related to this ion. More precisely, configuration interaction was retained among the following configurations: $3d^{10}$ + $3d^94s$ + $3d^95s$ + $3d^94d$ + $3d^95d$ + $3d^95g$ + $3d^84s^2$ + $3d^84p^2$ + $3d^84d^2$ + $3d^84s4d$ (even parity) and $3d^94p$ + $3d^95p$ + $3d^94f$ + $3d^95f$ + $3d^84s4p$ + $3d^84s4f$ + $3d^84p4d$ (odd parity). The core-polarization effects (CP) were introduced in the calculations according to a procedure described by Quinet *et al* [48] leading to the HFR + CP approach. For the dipole polarizability, α_d, we have adopted the value of 0.30 au corresponding to the ionic core $1s^22s^22p^63s^23p^6$. This value was derived from HFR oscillator strengths calculated in our previous work [43] for argon-like copper (Cu XII). The cut-off radius, r_c, was chosen equal to 0.762 au which corresponds to the HFR average value $\langle r\rangle$ for the outermost 3p core orbital. In addition, we have proceeded to a least-squares fitting of the calculated eigenvalues of the Hamiltonian to the experimental energy level values taken from [17, 18]. The adjusted parameters have been reported elsewhere [43] and will not be repeated here. For the Slater parameters not varied in the least-squares fit, a scaling factor of 0.90 was introduced [47]. The mean deviations reached 32 cm^{-1} for the even parity (72 levels, 24 adjusted parameters) and 103 cm^{-1} for the odd parity (120 levels, 26 parameters).

The HFR transition probabilities and lifetimes are reported in columns 6 and 7 of table 1.

5. Results and discussion

The experimental transition probabilities obtained in this work, as well as the HFR lifetime values and the lifetimes calculated by Theodosiou [38] (which agree quite well), are presented in table 1. The wavelengths of the investigated transitions, obtained from the energy levels reported in NIST tables [17, 18], and their relative intensities as determined in the present work, are also reported in the table (when $A_{ki} > 0.03 \times 10^7$ s^{-1}). The experimental relative transition probabilities have been normalized using the HFR radiative lifetimes of the upper energy levels.

In some cases, spectral lines observed in the present experiment (202.713, 218.937, 223.040, 226.457 and 237.630 nm) were blended by Cu I and Cu II spectral lines. As a consequence, the values of transition probabilities of the corresponding transitions, as well as for the weak line at 213.540 nm, were not directly measured on the spectra but were evaluated

Table 1. Transition probabilities (A_{ki}, in $10^7\ s^{-1}$) for $3d^9\ 4p$–$3d^9\ 4d$ and $3d^94p$–$3d^84s^2$ transitions of Cu II. The lifetime values of the upper levels (τ in ns) are also given. Only the transitions with $A_{ki} > 0.03 \times 10^7\ s^{-1}$ are quoted in the table.

Lower level	Upper level	λ (nm)[a]	A_{ki} (This work exp.)	A_{ki} (This work HFR)	τ (This work HFR)	τ (Previous)
4p $^3F^\circ_4$	4d $^3F_4$	209.840	20.91 ± 1.67	23.17	1.397	1.332[b]
4p $^1F^\circ_3$		219.568	42.00 ± 3.78	39.33		
4p $^3D^\circ_3$		224.897	9.11 ± 0.77	9.09		
4p $^3F^\circ_3$	4d $^3G_4$	211.731	72.0 ± 5.8	72.44	1.279	1.279[b]
4p $^3F^\circ_4$		213.009	4.4 ± 0.3	3.69		
4p $^1F^\circ_3$		223.040	2.02[c]	2.03		
4p $^3F^\circ_3$	4d $^1G_4$	202.219	1.14 ± 0.12	1.22	1.354	1.330[b]
4p $^3F^\circ_4$		203.384	0.89 ± 0.12	0.65		
4p $^1F^\circ_3$		212.511	22.83 ± 1.51	19.09		
4p $^3D^\circ_3$		217.498	49.00 ± 3.92	52.90		
4p $^3P^\circ_2$	4d $^1P_1$	196.736	1.80 ± 0.40	1.37	1.323	1.425[b]
4p $^3P^\circ_1$		202.713	12.73[c]	14.39		
4p $^3P^\circ_0$		206.626	20.00 ± 1.4	22.61		
4p $^3D^\circ_1$		226.537	12.46 ± 1.06	10.51		
4p $^3D^\circ_2$		227.834	13.15 ± 0.84	10.29		
4p $^1P^\circ_1$		229.100	17.57 ± 1.16	16.31		
4p $^3P^\circ_2$	4d $^3P_1$	202.995	7.99 ± 0.70	10.13	1.349	1.264[b]
4p $^3P^\circ_1$		209.364	22.00 ± 1.67	23.64		
4p $^3P^\circ_0$		213.540	0.43[c]	0.46		
4p $^3F^\circ_2$		218.286	8.02 ± 0.88	5.61		
4p $^1D^\circ_2$		226.321	15.37 ± 1.08	13.80		
4p $^3D^\circ_1$		234.873	2.82 ± 0.28	3.03		
4p $^3D^\circ_2$		236.268	0.63 ± 0.08	0.20[d]		
4p $^1P^\circ_1$		237.630	16.06[c]	17.25		
4p $^3P^\circ_2$	4d $^3P_2$	203.104	45.00 ± 3.60	52.70	1.283	1.257[b]
4p $^3P^\circ_1$		209.479	2.21 ± 0.24	1.97		
4p $^3F^\circ_3$		211.838	2.69 ± 0.26	2.25		
4p $^1F^\circ_3$		223.158	11.36 ± 0.91	10.80		
4p $^1D^\circ_2$		226.457	0.14[c]	0.17		
4p $^3D^\circ_3$		228.665	15.20 ± 0.90	9.36		
4p $^3D^\circ_2$		236.415	0.34 ± 0.03	0.42		
4p $^1P^\circ_1$		237.779	0.35 ± 0.13	0.13		
4p $^3P^\circ_2$	4d $^3S_1$	207.866	63.00 ± 4.41	65.57	1.242	1.250[b]
4p $^3P^\circ_1$		214.549	11.42 ± 0.96	11.83		
4p $^3P^\circ_0$		218.937	1.28[c]	1.33		
4p $^3D^\circ_1$		241.419	–	0.20		
4p $^3D^\circ_2$		242.893	1.93 ± 0.22	0.60		
4p $^1P^\circ_1$		244.333	2.00 ± 0.20	0.85		
4p $^3F^\circ_3$	$4s^2\ ^1G_4$	368.655	0.097 ± 0.010	0.108	380	5000[e], 2500[e]
4p $^1F^\circ_3$		404.348	0.114 ± 0.014	0.1177		
4p $^3D^\circ_3$		422.794	0.051 ± 0.004	0.0389		

[a] Deduced from the experimental energy levels compiled at NIST by Sugar and Musgrove [18].
[b] From [38].
[c] Blended transition. See the text.
[d] Cancellation effects present (CF < 0.01).
[e] From [44] (length and velocity forms).

from the theoretical estimates of the BFs. For that reason, no uncertainty is given in the table for the corresponding transition probabilities.

For most of the transitions, the total experimental uncertainties are ranging between 6% and 13% and include the statistical uncertainties as well as the errors due to calibration of the experimental device. For two spectral lines only (196.736 and 237.779 nm), the error is larger than 13%.

As can be seen from the table 1, for most of the investigated transitions, the experimental *A*-values are in fair agreement with the HFR results, the discrepancies reaching in most cases only a few (<10)%. For a few very weak transitions (242.893, 244.333, 236.268, 218.286 and 237.779 nm) however, larger discrepancies are observed and are due, on the one hand, to the difficulty of measuring accurately weak lines and, on the other hand, to the difficulty of calculating accurately the corresponding BFs which are very sensitive to the composition of the wavefunctions. For the transition at 228.665 nm, the discrepancy reaches about 50% and is more puzzling. No clear explanation has been found for explaining this discrepancy.

A large discrepancy is observed between the HFR radiative lifetime of the $4s^2\ ^1G_4$ excited level and the results published in [44]. The large difference observed between the length and velocity formalisms in [44] is an indication that correlation effects play an important role for this level and were probably not properly taken into account in the calculation of Blagoev *et al* [44].

The calculations of Goldsmith and Boxman [45] and of Theodosiou [38] were performed using the Coulomb approximation which does not treat electron configuration in an adequate manner. These results are expected to be inaccurate when strong configuration interaction is present.

The most extensive comparison is possible with the CIV3 results of Donnelly *et al* [40]. The comparison has been reported previously [43] and we will only recall the basic conclusions here. In the mean, the HFR + CP results differed from the CIV3 results (for the whole sample of transitions reported in [43]) by about 16%. Larger discrepancies were observed for some specific transitions which is not unexpected in view of the large mixing occurring for some states (see table 3 of [43]). They were due to the fact that no fine tuning was introduced in the calculations of [40] and that the degree of correlation introduced in different states (limited basically by the computer resources available) was depending upon the occupancy of the 3d subshell leading eventually to inaccurate *f*-values for the transitions with different 3d occupancy. Similar conclusions are valid for the restricted set of transitions retained in the present work. As our HFR + CP results agree well with the experimental data, they are expected to be the most accurate data available for this set of transitions.

6. Conclusions

A-values of transitions emitted from levels belonging to the $3d^94d$ and $3d^84s^2$ electronic configurations of Cu II have been measured for the first time with the LIBS method. For comparison, transition probabilities have been calculated by a HFR approach, taking relativistic effects and core-polarization effects into account. For most of the transitions, except a few very weak ones, good agreement is observed between experiment and theory. This agreement gives weight to the accuracy of this new set of transition probabilities in Cu II which is now established on an experimental basis. The present results are intended for guidance of future experimental investigations and to provide astrophysicists with some of the data they need for quantitative investigations of stellar spectra.

Acknowledgments

We acknowledge financial support from National Found of Bulgaria (grant 1516/05). Financial support from the Belgian National Fund for Scientific Research (FNRS) is also acknowledged. EB and PQ are Research Director and Research Associate of this organization.

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