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Feasibility of Using Boltzmann Plots to Evaluate the Stark Broadening Parameters of Cu(I) Lines

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Abstract

A linear Boltzmann plot was constructed using Cu(I) lines of well-known atomic parameters. Aligning other spectral lines to the plot was adopted as a viable way to estimate the most probable values of Stark broadening parameters of Cu(I) lines at 330.79, 359.91, and 360.2 nm. Plasma was generated by focusing neodymium-doped yttrium aluminum garnet (Nd:YAG) laser radiation at wavelength 532 nm on a pure copper target in open air. Plasma emission was recorded at delay times of 3, 4, 5, 7, and 10 μs. The in situ optically thin H_α line was used to determine the plasma reference electron density over the entire experiment. Following this method, the missing values of the Stark broadening parameters of the three Cu(I) lines turn out to be about 0.15 ± 0.05 Å (for 330.79 nm transition) and 0.17 ± 0.05 Å (for 359.91 360.20 nm transition) at reference electron density of $(1 \pm 0.09) \times 10^{17}\,cm^{-3}$ and temperature of $10\,800 \pm 630$ K. The apparent variation in plasma parameters at different delay times was found to scale with electron density and temperature as $\sim n_{e.} T_e^{0.166}$.

Keywords

Laser-induced breakdown spectroscopy, LIBS, Cu(I) spectral lines, Boltzmann plot method, Stark broadening coefficient, transition probability

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Introduction

The optical emission spectroscopy technique (OES) is a common technique used to diagnose plasmas produced by laser.[1–6] It was assumed that the emitted light is sufficiently influenced by plasma parameters (electron density and temperature) in addition to target sample ingredients.[5,6] The precision of measured values of plasma parameters depends on the accuracy of the available set of atomic constants (e.g., transition probability, upper-level energy, statistical weight, and Stark broadening parameters, etc.).[7] For plasmas in the state of local thermodynamic equilibrium (LTE), the measurement of the Stark component at full width half-maximum (FWHM) of an optically thin spectral line leads to accurate knowledge of the plasma genuine electron density and vice versa.[1–10] The natural process of plasma self-absorption effectively leads to distortion of emission line shape and consequently over and/or underestimated of plasma parameters.[9–14] However, the distorted lines can be recovered after carrying well-known procedures utilizing the presence of the optically thin H_α line.[9–14]

The Boltzmann plot method has been previously used to estimate the missing values of transition probabilities and Stark broadening parameters of several atomic transitions.[10,15–25] This should be ultimately carried out provided that at least one of these quantities should be available with sufficient accuracy.[1,5,6,8,10] Several authors calculated the Stark broadening parameters of several transition lines for the copper and some other elements using different techniques.[7,26–31]

In this paper, we shall confirm the feasibility of using the Boltzmann plot to estimate the missing values of Stark

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broadening parameters of the three Cu(I) lines at wavelength transitions of 330.79, 359.91, and 360.2 nm.

Experimental Setup

The experimental setup has been described in detail in previous publications.[9,12–14] It comprises a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser at a wavelength of 532 nm, pulse duration from 5 ns, and 100 ± 5 mJ output pulse energy. The plasma is created at the surface of flat a copper target (99.99% purity; K.J. Lasker Ltd) in the open air. The target was fixed on a homemade *x*,*y*,φ translational table holder to provide fresh target condition at each laser shot as shown in Fig. 1a. The laser beam was focused using an achromatic lens having $f_{\text{-number}} = 3.3$ while the target was positioned at a distance of 95 ± 2 mm from laser focusing lens to avoid breakdown in the air layer surrounding the target. The laser spot size with a diameter of 0.9 ± 0.1 mm was measured at the target surface using thermal paper. The plasma emission spectrum was brought to the entrance hole of the SE200 Echelle spectrograph using an optical fiber with a numerical aperture of 0.22 positioned at a distance of 7 ± 0.5 mm from the laser–plasma axis. In order to protect the tip of the optical fiber from the precipitations produced by the fast hot plasma species (which might cause an optical absorption window), we have used a thin quartz tube cap placed on the optical fiber tip. This home-fabricated protection cap was made so as it can be replaced from one experiment to another. The plasma evolution was traced at different ICCD camera delay times varying from 3 to 10 μs with a gate time of 1 μs over five laser shots on fresh target surface condition via rotating the target holder table. The spectral sensitivity of the camera–spectrograph–optical fiber system (Fig. 1b) was calibrated using an absolute radiometric lamp with correction factors (C_r) in the units of μW/count at each Cu(I) emission spectral line.[7,11] It is evaluated and given in Table I. The spectral line intensity was taken as the area under the curve.

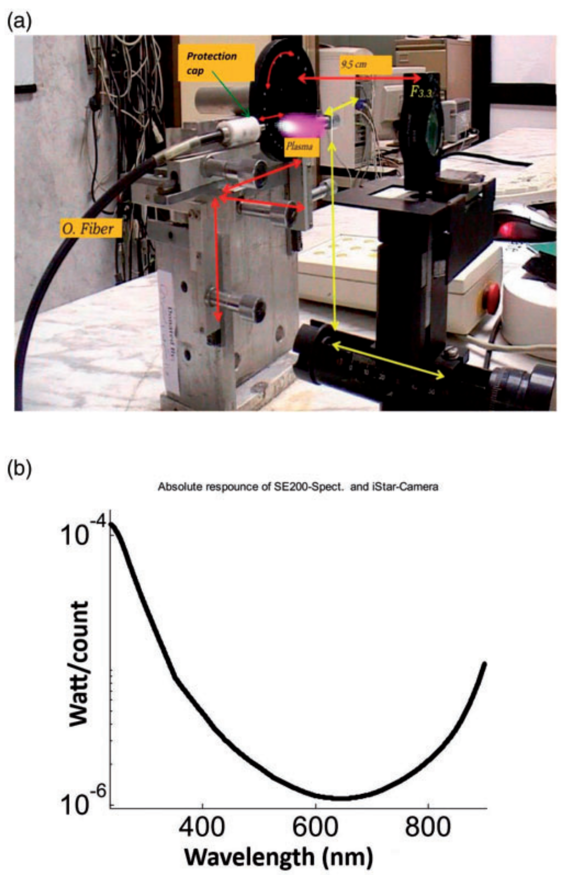

Figure 1. (a) A photograph of the homemade *x*,*y*,φ target holder and (b) the absolute spectral sensitivity of the measuring apparatus including optical fiber, SE200 spectrograph, and the ICCD camera.

Results and Discussion

The twelve Cu(I) lines of interest were identified as shown in Figs. 2a to 2l at wavelengths of 324.75, 327.39, 330.79, 359.91, 360.20, 427.51, 465.11, 510.55, 515.32, 521.82, 793.31, and 809.26 nm in addition to the H_α line at 656.27 nm. Nine lines at 324.75, 327.39, 427.51, 465.11, 510.55, 515.32, 521.82, 793.31, and 809.26 nm of well-known atomic parameters including transition probability and Stark broadening parameters (as given in Table II) were used to construct a standard Boltzmann plot (after carrying out the necessary examination against effect of self-absorption).[28,32–34] This straight line relation will be considered as a calibration graph in order to estimate the Stark broadening parameters of the Cu(I) lines at 330.79, 359.91, and 360.20 nm.

A glance at data in Table II concludes that the values of the Stark broadening parameters ω_S^{line} for the wavelengths 330.79, 359.91, and 360.2 nm transitions are absent.[28,32–34] Theoretically, these values can be calculated according to different theoretical approaches (impact theory and/or quasi-static approximation).[2,4,32–34] On the other hand, direct precise measurements of these quantities should be undertaken. In this context, we have adopted the following procedures in consequence including the fine analysis of spectral line shapes of Cu(I) lines emitted from plasma produced by laser.

On a par with these measurements, the good knowledge of the reference plasma electron density provides the basic millstone. Fortunately, the presence of the hydrogen alpha H_α line which appeared in emission spectra (predominantly from the very small concentration of humidity around the target surface in open air) can play this role.[9] Being optically thin, we have measured the plasma reference density utilizing the specific advantage of this line which stays a long time after plasma ignition.[9] Rigorous procedures were carried via fitting the spectral line shape of the H_α line to the Voigt function taking into account the instrumental

Table 1. Coefficients of spectral sensitivity at each Cu(I) line.

Wavelength (nm)	324.75	327.39	330.79	359.91	360.20	427.51	465.11	510.55	515.32	521.82	793.31	809.26
C_r (μW/count)	16.042	15.942	13.6940	7.7447	7.7447	3.4455	2.4457	1.7811	1.7198	1.6510	2.0297	2.3266
$\Delta\lambda^{inst}$ (nm)	0.1353	0.1364	0.1378	0.1500	0.1501	0.1781	0.1938	0.2127	0.2147	0.2174	0.3305	0.3372

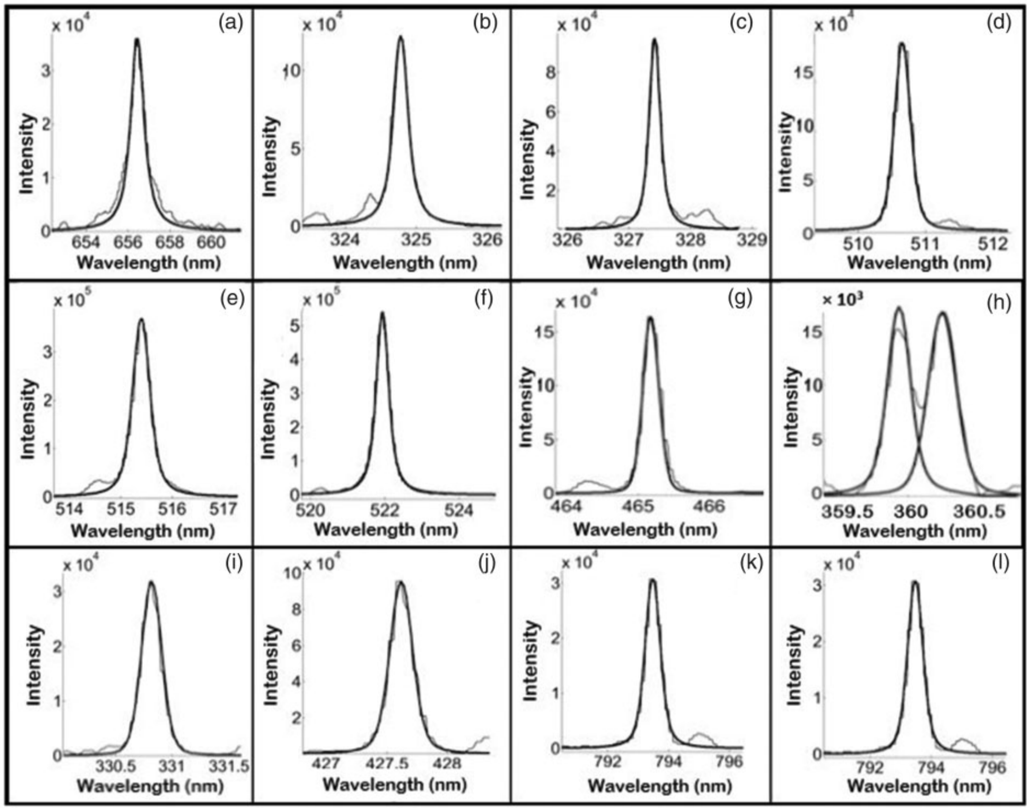

Figure 2. (a–l) Emission spectral lines of neutral copper at arbitrary delay of 4 μs, gate of 1 μs, and together with best fitting to Voigt function (thick solid lines).

bandwidth at this wavelength (0.36 nm). Table III lists measured plasma reference density at different delay times featuring a regular monotonic decline.

Next, we checked the existence of any distortion to the recorded Cu(I) spectral lines via plasma self-absorption using calculation of the coefficients of plasma self-absorption SA^{line}. This factor tends to be larger than zero and equal to one in the limit of purely optically thin line.[9,12–14] This coefficient is given by the following expression

$$SA^{line} = \left(\frac{n_e^{line}}{n_e^{H_\alpha}}\right)^{-0.54} \quad (1)$$

where n_e^{line} is the electron density of any of the nine Cu(I) lines as calculated from the following expression

$$n_e^{line} = \Delta\lambda_s^{line}\left(\frac{N_r^*}{\omega_S^{line}}\right) \quad (2)$$

The measured Lorentzian FWHM of each of the nine Cu(I) lines $\Delta\lambda_s^{line}$ and the Stark broadening parameters ω_S^{line} at certain specific reference electron density N_r^* are listed in Table II.[28,34]

This Lorentzian FWHM $\Delta\lambda_s^{line}$ was carefully extracted from the experimentally measured line shape via fitting to the theoretically constructed Voigt function taking into account the instrumental component (of a Gaussian nature) $\Delta\lambda_{instrum}$. The values of the calculated instrumental bandwidths at different transition wavelengths are presented in Table I. We have neglected other broadening mechanisms, e.g., Van der Wall broadening, which was found to contribute by nearly 0.01 nm, is important in cases of relatively low temperature cases (~0.3 eV). Also, the resonance broadening was found to contribute by nearly 3% to the line shape width.[3,35] The results of the fitting procedure are shown in Figs. 2a to 2l.

Table II. Atomic parameters of the identified Cu(I) lines together with their respective Stark broadening parameters and transition probabilities and the accuracy of each as given at different references.[28,32–34]

λ (nm)	Transition lower	Transition upper	Trans. probability A (s^{-1})	Acc. $\frac{\Delta A}{A}$	Ref. for A	g	E_{exc} (eV)	Stark broadening ω_S (nm)/reference density N_r^* (cm^{-3})	Acc. $\frac{\Delta\omega_S}{\omega_S}$	Ref. for ω_S
324.75*	$3d^{10}4s$	$3d^{10}4p$	1.39×10^8	5%	32	4	3.82	$0.000301/10^{16}$	35%	28
327.39*	$3d^{10}4s$	$3d^{10}4p$	1.37×10^8	5%	32	2	3.78	$0.000301/10^{16}$	35%	28
330.79	$3d^9(^2D)\ 4s4p(^3P^\circ)$	$3d^94s\ (^3D)\ 4d$	2.21×10^8	–	33	12	8.82	–	–	–
359.91	$3d^9(^2D)\ 4s4p(^3P^\circ)$	$3d^9\ 4s(^3D)\ 4d$	1.30×10^8	–	33	10	8.83	–	–	–
360.21	$3d^9(^2D)\ 4s4p(^3P^\circ)$	$3d^9\ 4s(^3D)\ 4d$	1.60×10^8	–	33	8	8.83	–	–	–
427.51	$3d^9(^2D)\ 4s4p(^3P^\circ)$	$3d^9\ 4s(^3D)\ 5s$	3.50×10^7	25%	32	8	7.73	$0.080/10^{17}$	50%	34
465.11	$3d^9(^2D)\ 4s4p(^3P^\circ)$	$3d^9\ 4s(^3D)\ 5s$	3.80×10^7	25%	32	8	7.73	$0.087/10^{17}$	50%	34
510.55	$3d^94s^2$	$3d^{10}4p$	2.00×10^6	20%	32	4	3.81	$0.043/10^{17}$	50%	34
515.32	$3d^9(^2D)\ 4s4p(^3P^\circ)$	$3d^94s(^3D)\ 5s$	6.00×10^7	20%	32	4	6.19	$0.190/10^{17}$	50%	34
521.82	$3d^{10}4p$	$3d^{10}4d$	7.50×10^7	20%	32	6	6.19	$0.220/10^{17}$	50%	34
793.31	$3d^{10}4p$	$3d^{10}5s$	2.24×10^7	–	33	2	5.34	$0.320/10^{17}$	50%	34
809.26	$3d^{10}4p$	$3d^{10}5s$	4.60×10^7	–	33	2	5.34	$0.240/10^{17}$	50%	34

* Indicates the resonance transition.

Table III. Plasma reference electron density measured utilizing the H_α line at different delay times.

Delay time (μs)	3	4	5	7	10
$n_e^{H_\alpha}(cm^{-3})$	9.60×10^{16}	5.00×10^{16}	3.10×10^{16}	1.96×10^{16}	6.30×10^{15}

The SA coefficients inherent to the nine Cu(I) lines have been calculated and presented in Table IV. It is worth noting that the spectral lines at 427.51 and 465.11 nm reveal SA coefficients which are close to unity at an early delay time of 3 μs. This means that these lines can be considered as initially optically thin before being opaque to plasma radiation reabsorption at later stage of plasma expansion. In contrast, the resonance transition lines at 324.75, 327.39 nm show very small SA values ranging from around 10^{-4} down to 10^{-5}. Hence, these lines should be subjected to careful correction against self-absorption as being optically thick. Meanwhile, a glance at Table IV shows that other Cu(I) lines at 510.55, 515.32, 521.82, 793.31, and 809.26 nm appear to undergo a moderate amount of self-absorption with SA decreasing from around 0.3 down to 10^{-2} with delay time. Obviously, as the delay time increases, the plasma is naturally cooling down via adiabatic expansion, and hence the larger the probability for the emitted photons to be reabsorbed.[3,26,34]

The following expression is used to correct (recover) the measured (distorted) spectral intensity $\bar{I}^{line}_{exp}$ through coefficients of self-absorption, i.e.,[7,9–12]

$$\bar{I}^{line} = \frac{\bar{I}^{line}_{exp}}{(SA^{line})^{0.46}} \tag{3}$$

where the symbol ($\bar{I}^{line}$) indicates the spectral line intensity measured as the area under the curve instead of peak spectral radiance. Moreover, this corrected spectral intensity $\bar{I}^{line}$ should nonetheless be corrected by the coefficients of spectral sensitivity of the used instrument, including optical fiber, spectrograph, and intensified charge-coupled device (ICCD) camera C_r as given in Table 1 and Fig. 1b. Thereupon, we have Eq. 4

$$\bar{I}^{line}_0 = \bar{I}^{line} \times C_r \tag{4}$$

Finally, the Boltzmann plot of the quantity defined by $\ln\left(\bar{I}^{line}_0 \lambda_{line}/g^{line}A^{line}\right)$ versus excitation energy E^{line}_{exc}(eV) was constructed. Ultimately, it proves to yield a perfect straight line of negative slope as shown in Fig. 3 taken at delay time (T_d) of 4 μs. It incorporates the nine spectrally corrected spectral radiance Cu(I) lines of well-known transition probability and Stark broadening parameters. It is worth noting that the slope of this straight line is the electron temperature which is around $T_e = 9000$ K.[36]

As a final step, we have utilized the linearity of the Boltzmann plot (shown in Fig. 3) to estimate the most probable values of the Stark broadening parameters at three wavelengths of 330.79 nm (^{4}G–4 F°) transition, 359.91 nm for (^{4}F–4D°) transition, and 360.20 nm for the (^{4}D–4D°) transition (marked by a green cross).

Table IV. Calculated coefficients of self-absorption SA of the Cu(I) lines of known transition probability at different delay times.

	Coefficient of self-absorption (SA) of Cu(I) lines								
Delay time (µs)	324.75* (nm)	327.39* (nm)	427.51 (nm)	465.11 (nm)	510.55 (nm)	515.32 (nm)	521.82 (nm)	793.31 (nm)	809.26 (nm)
3	0.000671	0.0012	0.6180	0.95	0.2959	0.4873	0.3567	0.5711	0.6380
4	0.000114	0.00024	0.4763	0.88	0.0586	0.2323	0.1789	0.2584	0.2676
5	0.000044	0.000091	0.6636	0.83	0.0275	0.2179	0.1416	0.2706	0.1834
7	0.000015	0.000023	0.3007	0.7441	0.0017	0.0672	0.0641	0.0390	0.0371
10	0.000004	0.000005	0.2811	0.3283	0.0019	0.0238	0.0257	0.0150	0.0133

* Indicates the resonance transition.

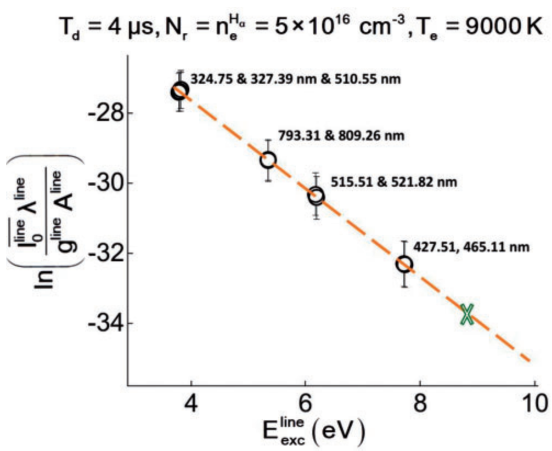

Figure 3. Boltzmann plot constructed at delay time 4 µs using nine well-known transition probabilities and Stark broadening parameters of Cu(I) lines at 324.75, 327.39, 427.51, 465.11, 510.55, 515.32, 521.82, 793.31, and 809.26 nm (open circles) with green cross indicating the appropriate location of the unknown Stark broadening parameters of transition at 330.79, 359.91, and 360.2 nm.

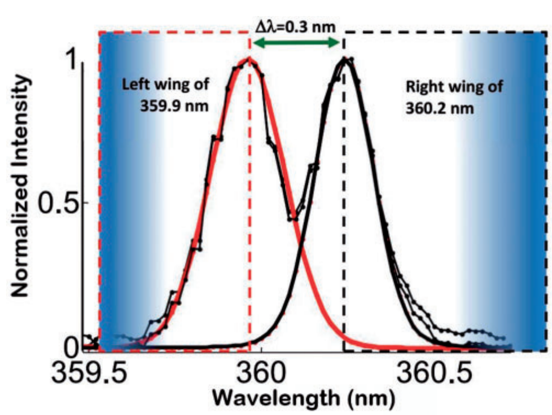

Figure 4. Voigt fitting (red curve at 359.9 nm and black curve at 360.2 nm) to spectral line data (black dotted curve) showing the wings (dashed squares) used to evaluate electron density from both lines at wavelengths 359.91 (unhighlighted left) and 360.2 nm (unhighlighted right) at arbitrary delay time of 5 µs.

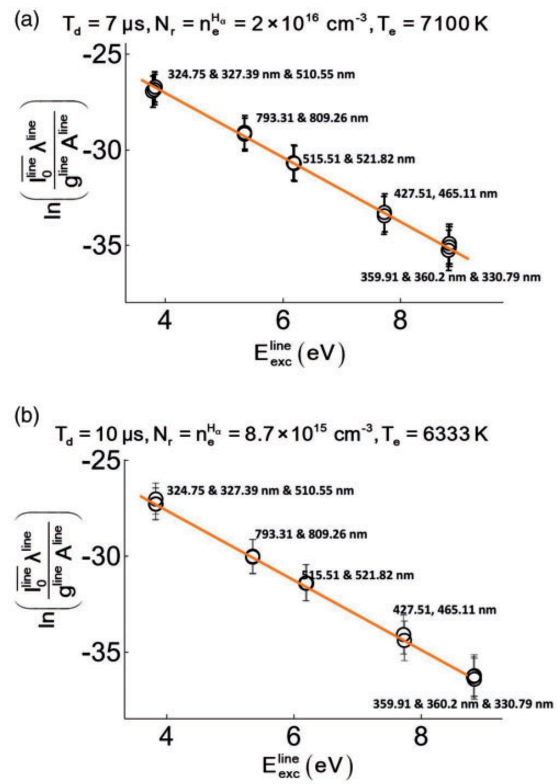

Figure 5. Boltzmann plots at different delay times (a) 7 µs and (b) 10 µs with values corresponding to three Cu(I) transition lines (at the far right side of the graph) coincide with linear plot.

Traditionally, we start with fitting the experimentally measure lines profiles to Voigt line shape using the Stark broadening parameters of the three investigated lines as fitting parameters until the best fitting of the values $\ln\left(\overline{I_0^{line}}\lambda_{line}/g^{line}A^{line}\right)$ to the Boltzmann line is achieved. The software eventually terminates with the output of the estimated parameters as shown in Figs. 5a and 5b.

One major problem that should be fixed concerns the limited resolving power of our spectrograph causing an instrumental bandwidth of 0.14 nm in the near UV region. This would consequently affect the resolution of the two Cu(I) lines at 359.91 and 360.20 nm which are close by

Table V. Final results of the estimated Stark broadening parameters of the three Cu(I) lines at different reference density and temperatures.

Delay time (μs)	Reference electron temperature (K)	Reference electron density $N_r = n_e^{H_\alpha}$ (cm^{-3})	Estimated Stark broadening parameter of Cu(I) lines (nm)			Average Accuracy $\frac{\Delta\omega_S}{\omega_S}$
			ω_S (330.79 nm)	ω_S (359.91 nm)	ω_S (360.20 nm)	
3	10,801	9.60×10^{16}	0.0150 ± 0.0078	0.0170 ± 0.0088	0.0170 ± 0.0088	51.9%
4	9152	5.00×10^{16}	0.0110 ± 0.0050	0.0130 ± 0.0060	0.0130 ± 0.0060	51.9%
5	7500	3.10×10^{16}	0.0042 ± 0.0021	0.0048 ± 0.0024	0.0048 ± 0.0024	51.9%
7	6728	1.96×10^{16}	0.0032 ± 0.0017	0.0032 ± 0.0016	0.0032 ± 0.0016	51.9%
10	6333	6.30×10^{15}	0.0011 ± 0.0005	0.0012 ± 0.0006	0.0012 ± 0.0006	51.9%

0.3 nm. This problem has been overridden by utilizing the symmetric nature of the Voigt function which requires only one clear wing of the line under consideration to reach good fitting (unhighlighted left part of the line at 359.9 nm and unhighlighted right part of the 360.2 nm) as shown by the shaded areas in Fig. 4.[2,3] This technique enabled us to just double the extracted half-width of the spectral lines at 359.9 nm and 360.2 nm and to get the Voigt FWHM hence the electron density.

This procedure was repeated at different delay times as demonstrated in Figs. 5a and 5b, whereby the fitting value of Stark broadening parameters of the three transitions could be deduced.

Table V summarizes the final results of the estimated Stark broadening of the Cu(I) transition lines at 330.79, 359.9, and 360.2 nm at different delay times (i.e., at different values of plasma parameters). The calculated average relative accuracy of about 52% is determined by adopting the method reported by Koshelev et al.[7] This uncertainty arises mainly from the available Stark broadening parameters of spectral lines transitions at wavelengths ranging from 427.51 to 809.11 nm at 50% and in best cases of 35% for the resonance transitions (as indicated in Table II).[28] In addition, uncertainty in the available values of the transition probabilities of the respective Cu(I) lines is found to vary from 5% (class AAA) in the case of resonance lines up to 25% for other lines as indicated in Table II.

With no available standard data to contrast our experimental findings, the trend, however, of our estimated values at different plasma parameters was nevertheless investigated in the light of the theoretical approach conducted by Zmerli et al.[28]

Figures 6a and 6b corroborate the scaling of predicted Stark broadening parameter with electron temperature as $\omega_s^{Transition} \alpha n_e \cdot T_e^{0.16667}$ for the three lines of interest at 330.79, 359.91, and 360.20 nm. As can be clearly seen, the excellent agreement with theoretical prediction would indicate that the experimental findings are fairly reliable.

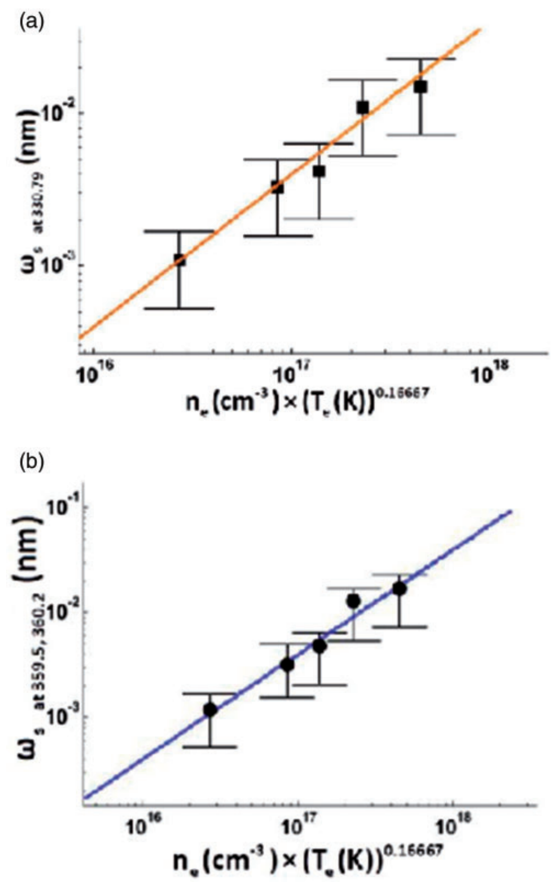

Figure 6. Variation of the estimated Stark broadening parameters of the (a) Cu(I) line at 330.79 nm and (b) the two lines at 359.91, 360.2 nm with plasma parameters (density and temperature).

Conclusion

We have measured the Stark broadening parameters of three Cu(I) lines at 330.79, 359.91, and 360.20 nm at different reference electron densities in the range from 1×10^{17} down to 8×10^{15} and temperatures of nearly 10 800 to

6350 K using the Boltzmann plot method. Stark broadening parameters of 0.15 ± 0.05 Å at the 330.79 nm (3d^{9}4s (^{3}D) 4 d (^{4}G–4 F°)) transition and 0.17 ± 0.05 Å at the transitions (^{4}F–4D°) at 359.91, and (^{4}D–4D°) at 360.20 nm have been estimated at reference electron density of 1×10^{17} cm^{-3} and temperature of 10,800 ± 630 K. These parameters are found to vary in a monotonic manner with plasma parameters as $\omega_s^{Transition} \alpha n_e \cdot T_e^{0.16667}$. We are looking forward to establishing this method as a reliable technique for the measurement of transition probability as well as Stark broadening parameters of unknown transitions.

Declaration of Conflicting Interests

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