Observation of density-enhanced dielectronic satellite spectra produced during subpicosecond laser-matter interactions

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An experimental verification of the theoretically predicted density enhancement of the dielectronic satellite spectra emitted by multiply charged lithiumlike ions in high-density plasmas is reported. These plasmas were created by the interaction of solid MgF_2 and SiO_2 targets with high-intensity, subpicosecond, ultraviolet laser radiation. Comparisons between the observed and theoretically predicted values for the intensity ratios between two different pairs of dielectronic satellite lines have enabled determinations of electron densities of the order of 10^{23} cm^{-3} to be made in these high-density laser-produced plasmas.

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I. INTRODUCTION

Recently developed [1] high-power, subpicosecond laser sources have been used in the creation of high-density plasmas from various solid targets for the high-resolution study of x-ray emission spectra from multiply charged ions [2], for the investigation of Stark broadening [3], and for the examination of nonlinear-optical phenomena [4]. Since these high-intensity laser sources give rise to strong electromagnetic fields ($\sim 0.1-10e/a_0^2$), multiphonon excitation and ionization processes are expected to play an important role in the initial stages of the plasma formation. Furthermore, strong-field phenomena may also influence certain characteristics of the spectrum of the emitted radiation during the intense laser-matter interaction [5].

In order to conduct a detailed investigation of the processes of ionization and the production of x-ray emission during the high intensity laser-matter interaction, it is necessary to develop spectroscopic methods for the analysis of high-temperature and high-density plasmas. In this investigation, we report experimental verification of the theoretically predicted [6] density enhancement of the dielectronic satellite spectra emitted by multiply ionized lithiumlike ions in high-density plasmas. These plasmas were created by the interaction of various solid targets with high-intensity, subpicosecond (~ 600 fs), ultraviolet (KrF*, 248 nm) laser radiation. This unique spectral phenomenon, which arises from the collisionally induced angular momentum redistribution of the population among autoionizing states undergoing spontaneous radiatively stabilizing transitions, has enabled the direct determination of the electron density generated in these high-density laser-produced plasmas.

The dielectronic satellites of interest are situated on the long-wavelength side of the resonance lines emitted by the multiply charged heliumlike ions and are produced by spontaneous radiatively stabilizing transitions, with rates denoted by $A_r(a \to f)$, from autoionizing states "a" to bound states "f" of the corresponding lithiumlike species. In the low-density corona-model theory of the satellite-line intensities [7,8], one assumes that the autoionizing states "a" are populated either by the inner-shell-electron collision excitation process from a lithiumlike bound state "b," whose rate per unit electron density N_e is denoted by $C_e(b \to a)$, or by the radiationless electron-capture process from a heliumlike state "i," whose rate per unit electron density is denoted by $C_{\text{cap}}(i\epsilon_i \to a)$. The radiationless electron-capture process is the initial step in the two-step mechanism of dielectronic recombination (DR) described by Burgess [9]. Accordingly, the total intensity $I(a \to f)$, defined as the rate of spontaneous radiative emission per unit volume in the dielectronic satellite transition $a \to f$, may be expressed in the form

$$I(a \to f) = \sum_i \alpha_{\text{DR}}(i\epsilon_i \to a \to f)N(i)N_e + \sum_b C_R(b \to a \to f)N(b)N_e \ , \tag{1}$$

where the densities $N(i)$ and $N(b)$ refer to recombining ions initially in the state "i" and recombined ions in the state "b", respectively. However, it has been demonstrated by Jacobs and Blaha [6] that, with increasing plasma density, the populations of the autoionizing states can be substantially altered from their low-density values by collisional transitions among the autoionizing states. The rate coefficients $\alpha_{\text{DR}}(i\epsilon_i \to a \to f)$ and $C_R(b \to a \to f)$ describing the emission of dielectronic-recombination radiation and radiation produced by inner-shell-electron collisional excitation, respectively, can be written in the generalized forms [6]

$$\alpha_{\text{DR}}(i\epsilon_i \to a \to f) = \sum_{a'} A_r(a \to f)Q^{-1}(a,a') \times C_{\text{cap}}(i\epsilon_i \to a') \ , \tag{2}$$

$$C_R(b \rightarrow a \rightarrow f) = \sum_{a'} A_r(a \rightarrow f) Q^{-1}(a,a') C_e(b \rightarrow a') . \quad (3)$$

In the low-density corona-model approximation, the matrix Q^{-1} has only diagonal elements, which are given by the sums of the autoionization and spontaneous radiative decay rates. With the inclusion of collisional transitions among the autoionizing states, it is necessary to invert the generalized collisional-radiative transition matrix Q, in which transition rates among the autoionizing states occur in both the diagonal and off-diagonal elements, as described by Jacobs and Blaha [6].

In particular, angular-momentum-changing electron-ion collisions are very effective in redistributing population among the autoionizing states. Moreover, since these angular-momentum-changing collisional transitions are usually the most rapid electron-ion collisional processes in a high-temperature plasma, they are likely, at sufficiently high density, to establish a local statistical-equilibrium distribution of populations among the autoionizing states. The collisionally induced modification in the populations is especially significant for the metastable autoionizing states, since they have relatively small autoionization and radiationless electron-capture rates. This modified distribution of populations among the autoionizing states gives rise to a dramatic density enhancement of the dielectronic satellite spectra in the high-density region.

The sets of dielectronic satellite lines under investigation are, following the notation of Gabriel [7] for the various fine-structure components, the $1s2p^2\,{}^2D \rightarrow 1s^2 2p\,{}^2P$ (j,k,l), $1s2p^2\,{}^2P \rightarrow 1s^2 2p\,{}^2P$ (a,b,c,d), and the $1s2p({}^1P)2s\,{}^2P \rightarrow 1s^2 2s\,{}^2S$ (q,r) transitions. An analysis based on the total intensities that are defined as sums over the fine-structure components is appropriate for the ions considered in the present investigation, because the fine-structure splittings of their satellite lines are not significant. The intensity of the (j,k,l) satellite lines, which depends predominantly on the He-like ion abundance, provides a convenient reference, because it has been predicted [6] to vary quite weakly with electron density. On the other hand, the intensity of the (a,b,c,d) satellite lines, which is vanishingly weak at low densities where their formation occurs predominantly as a result of inner-shell-electron collisional excitation from the less-abundant Li-like ion, exhibits a dramatic enhancement [6] at high plasma densities, due to the process of dielectronic recombination induced by collisional transitions among the autoionizing states. For ions with nuclear charges Z in the range from 12 to 14, this collisional mixing effect is significant at electron densities N_e in the range from 10^{22} to 10^{24} cm^{-3}. This range of electron density is of particular interest in the present study, because it spans the region from the critical electron density for 248-nm laser radiation to solid electron density. Furthermore, it should be emphasized that, since the radiating ions of interest are significantly abundant only for a relatively short time before plasma cooling and expansion can occur, the observation of the dielectronic satellite lines provides a diagnostic snap shot of the state of this hot high-electron-density laser-produced plasma.

The remainder of this paper is organized in the following manner. In Sec. II, the experimental investigation is described, including the characteristics of the laser system, the methods of spectral intensity measurement, and the physical properties of the laser-produced plasmas. The determination of the electron density, based on the comparisons between the observed and theoretically predicted values for intensity ratios involving the two different sets of dielectronic satellite lines, is described in Sec. III. We provide discussions of both the limitations inherent in the spectral observations and the validity of the theoretical assumptions, and a reasonable case is made that the reported electron density follows from a comparison between the experimental results and the theoretical predictions. Finally, our conclusions are presented in Sec. IV.

II. EXPERIMENTAL INVESTIGATION

The KrF* (248 nm) laser used in our experimental investigation has a pulse duration of $\sim$600 fs and a pulse energy of $\sim$0.15 J. This subpicosecond energy is accompanied by a poorly focusable (focal area $\sim 8\times10^{-2}$ cm^2) prepulse, having a duration of $\sim$20 ns and an energy of $\sim$10 mJ, which arises from the amplified spontaneous emission produced in the final amplifier of the laser system. A detailed description of this laser system together with its operational characteristics has been presented elsewhere [10]. The laser beam was focused with a CaF_2 lens onto flat targets, composed of either SiO_2 or MgF_2, and intensities up to $\sim 10^{17}$ W/cm^2 were produced at the target surfaces. High-resolution x-ray spectra were recorded on DEF film by flat potassium acid phthalate (KAP, $2d=26.6$ Å) and pentaerythritol (PET, $2d=8.7$ Å) crystal spectrometers. Each spectrum was produced by an integrated exposure from 25 laser shorts.

A question of particular concern in this work is the influence of a low-density plasma potentially produced by the prepulse, since it could alter the interaction of the subpicosecond pulse with the dense target material. Independent measurements have shown, however, that for the uv-transparent MgF_2 and SiO_2 targets, only the high-intensity subpicosecond laser energy interacts with the solid material [11].

The measured x-ray spectra exhibit several salient features. In particular, from the maximum principal quantum number observed in the He-like Mg and Si spectra, which may be deduced from the termination of the Rydberg series due to the phenomenon of continuum lowering, the electron-temperature–to–density ratio T_e/N_e in the emitting spatial region is estimated to be 125×10^{-23} eV cm^3. When referring to spectral observation, it is usually considered to be more correct to associate the maximum principal quantum number with an alternative phenomenon involving the advance of the series limit due to line merging rather than with the thermodynamic correction due to continuum lowering. However, the traditional approach to the description of the line-merging phenomenon is based on the quasistatic approximation for the electric fields produced by both electrons and ions, and this approximation may not be valid for the ultrashort-pulse laser-matter interactions that are the subject of this investigation. The states of these ions with the estimated maximum principal quantum numbers have radiative lifetimes in the range of a few picoseconds, only severalfold larger than the irradiating laser pulse itself. Furthermore, the actual lifetimes of the excited levels are expected to be reduced by collisional quenching. Moreover, due to the low rates of excitation characteristic of H-like $2p \rightarrow 1s$ transitions, the presence of $L\alpha$ emissions in the spectra of Mg and Si indicates that the plasma electron density N_e is at least in the range from 2×10^{22} to 5×10^{22} cm^{-3} and that the corresponding electron temperature T_e is between 500 and 1000 eV. Finally, because of the lower nuclear charge associated with the H- and He-like F transitions, these ions are expected to reflect correspondingly lower ranges of both density and temperature than the Mg and Si species.

The dependences of the intensity ratios of the dielectronic satellite transitions on changes in the electron temperature have been experimentally investigated by varying the intensity of irradiation for the MgF_2 and SiO_2 targets over a factor of ~ 30 and making the reasonable assumption that this leads to a corresponding variation of the electron temperature in the resulting plasma. The x-ray spectra obtained by means of this procedure are shown in Fig. 1. No substantial variations can be seen in the relative intensities among the various satellite lines. In contrast, with the same variation of focused laser intensities, the intensity ratios exhibited by the hydrogenlike $2p \rightarrow 1s$ and the heliumlike $1s3p \rightarrow 1s^2$ resonance lines of Mg and Si are found to change by approximately a factor of 10. This observed behavior of the resonance lines reveals the existence of a strong electron-temperature variation within the emitting plasma. However, the contrasting behavior of the satellite transitions is in agreement with the previous calculations reported by Jacobs and Blaha [6], which predict that the relative intensities of these dielectronic satellite transitions are weakly dependent upon the electron temperature. We note that this behavior also differs sharply from that shown by the intensities of the satellite lines when considered relative to the intensity of the associated resonance line emitted by the heliumlike ion. In this latter case, the ratios have been predicted to show strong electron-temperature dependences [6–8].

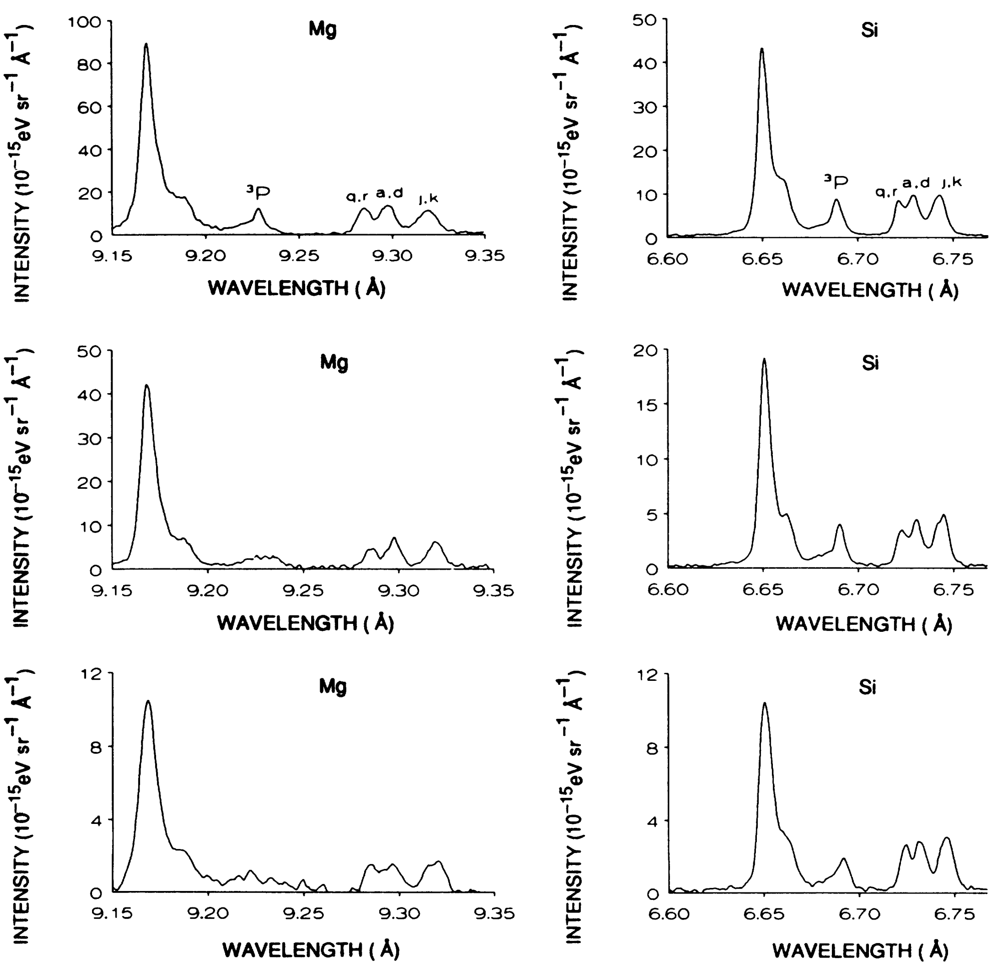

FIG. 1. Dielectronic satellite spectra associated with the $1s2p\ ^1P \rightarrow 1s^2\ ^1S$ resonance lines of heliumlike Mg and Si, obtained for various focused laser intensities. The alphabetical designation of the satellite transitions has been introduced by Gabriel [7].

It should also be noted that the ratio of the dielectronic satellite line intensity relative to that of the $1s2p\ ^1P \rightarrow 1s^2\ ^1S$ resonance line, which has been traditionally employed in the analysis of the spectra from low-density plasmas, is not expected to provide a precise determination of the local conditions within a very-dense plasma, since this resonance-line transition is the most susceptible in the spectrum to the effects of optical density. However, since the high-density plasmas created in the present study by high-intensity irradiation have maximum depths of only a few thousand angstroms [11], the majority of the x-ray transitions, which are radiated primarily from the high-temperature regions, can be regarded as optically thin.

In particular, these plasmas can be considered to be optically thin for emissions from the lithiumlike dielectronic satellite transitions of interest in the present investigation. The absorption coefficient $\alpha(f \rightarrow a,\omega)$, corresponding to the inverse of the photon mean free path for self-absorption of the dielectronic satellite transition $a \rightarrow f$ at the frequency ω, can be expressed, in terms of the population density $N(f)$ in the lower level "f" and the spontaneous radiatively stabilizing transition rate $A_r(a \rightarrow f)$, in the form [6]

$$\alpha(f \rightarrow a,\omega) = (\pi c/\omega)^2 A_r(a \rightarrow f) L(f \rightarrow a,\omega)(g_a/g_f) N(f) , \quad (4)$$

where g_a and g_f denote the statistical weights of the upper and lower levels, respectively, and $L(f \rightarrow a,\omega)$ is the frequency-normalized line-shape function. The optical depths associated with the relevant dielectronic satellite transitions for frequencies at the line centers are estimated to be of the order of 0.1, taking into account only thermal Doppler broadening and assuming that the emissions occur from a plasma with an ion temperature of 0.5 keV, an electron density of 5.0×10^{23} cm^{-3}, and a path length of 2500 Å. The actual optical depths of the dielectronic satellite transitions are expected to be further reduced from these estimates when Stark broadening is taken into account. Accordingly, it will not be necessary to treat radiation transport for the analysis of the dielectronic satellite spectra obtained in this experimental investigation.

III. ELECTRON-DENSITY DETERMINATIONS FROM THE INTENSITY RATIOS BETWEEN PAIRS OF DIELECTRONIC SATELLITE LINES

The theoretical predictions [6] for the dielectronic satellite intensities have been made under the assumption that the electron-ion encounters involve electrons with a Maxwellian velocity distribution, which enters into the evaluation of the various collisional transition rate coefficients in Eqs. (2) and (3). In the general case the electron velocity distribution must be determined from the time-dependent balance between the laser photon absorption processes and the various electron collisional and radiative emission processes. It has also been assumed that the autoionizing-state populations and charge-state distributions, which occur in the expression given by Eq. (1) for the dielectronic satellite line intensities, are determined by a time-independent, equilibrium balance established among the various elementary atomic collisional and radiative processes. A consideration of the relevant collisional and radiative time scales, similar to that presented by Cobble *et al.* [2], reveals that electron-electron collisions are sufficiently rapid, even for subpicosecond irradiation, to justify the assumption that the plasma electrons have attained a Maxwellian velocity distribution. However, the electron-ion collision times are generally most sufficiently short to satisfy the requirements for the establishment of the corresponding equilibrium values for the bound-state densities and charge-state distributions. The *exception* to this statement for the ion states occurs in the case of the angular-momentum-changing electron-ion collisions that are responsible for the density enhancement of the dielectronic satellite intensities. These electron-ion collision rates are sufficiently rapid to fulfill the conditions for the establishment of a local statistical-equilibrium distribution of populations among the autoionizing states. Since the density enhancement of the dielectronic satellite spectra is determined primarily by the redistribution of population among the autoionizing states, it will not be necessary to attempt a solution for the immensely more complex and uncertain time-dependent excitation and ionization balance problem in this investigation.

The radiative emission rate coefficients for the (j,k,l) dielectronic satellite lines have been predicted [6] to be relatively insensitive to the electron density. Consequently, these dielectronic satellite lines can serve as a suitable reference for comparison with the density-sensitive (a,b,c,d) dielectronic satellite lines. Furthermore, the predominant contributions to the intensities of both the (a,b,c,d) and the (j,k,l) dielectronic satellite lines depend at high densities only on the concentration of the He-like ion. Specifically for electron densities N_e greater than 1.0×10^{23} cm^{-3}, the dielectronic-recombination contributions from the He-like ion exceed by almost two orders of magnitude the contributions associated with inner-shell-electron collisional excitation of the Li-like ion, when angular-momentum-changing electron-induced collisional transitions among the autoionizing states are taken into account. Consequently, the enhancement of the intensity ratio $I(a,b,c,d)/I(j,k,l)$, which occurs at high densities, is not expected to be sensitive to the relative abundance of the He- and Li-like ions, and the corrections to the steady-state approximation are not expected to be large.

The dielectronic satellite intensity ratio $I(a,b,c,d)/I(k,k,l)$ is observed to be 0.9 for both Mg and Si. In addition, the dielectronic satellite intensity ratio $I(q,r)/I(j,k,l)$ is observed to be between 0.7 and 0.8 for both Mg and Si. These two observed dielectronic satellite intensity ratios represent substantial enhancements in comparison with the low-density corona-model predictions [7,8]. Comparisons with the theoretical findings [6] obtained for these particular satellite-line intensity ratios, which are reproduced for Si in Figs. 2 and 3, both lead to the result that the electron densities are of the order of

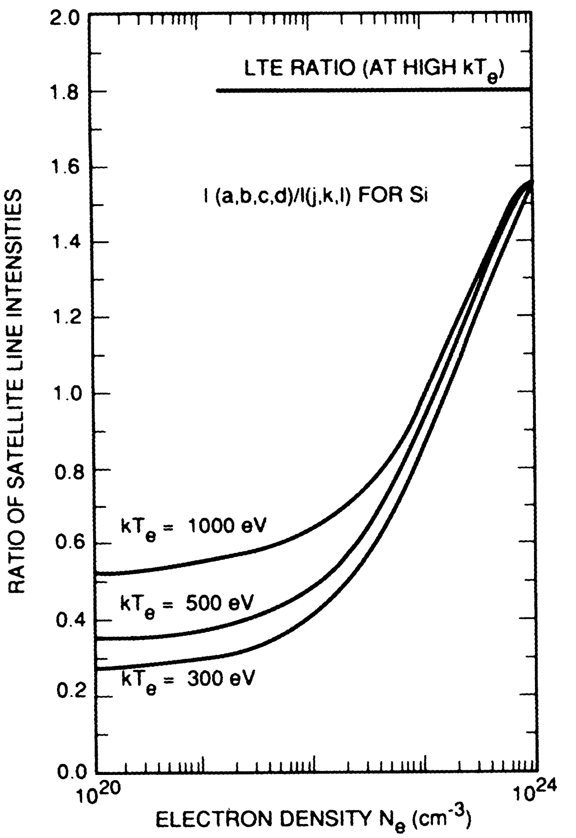

FIG. 2. The intensity ratio between the $1s2p^2\,{}^2P \rightarrow 1s^2 2p\,{}^2P$ (a,b,c,d) satellite lines and the $1s2p^2\,{}^2D \rightarrow 1s^2p\,{}^2P$ (j,k,l) satellite lines for Si, as a function of the electron density N_e for various values of the electron temperature T_e.

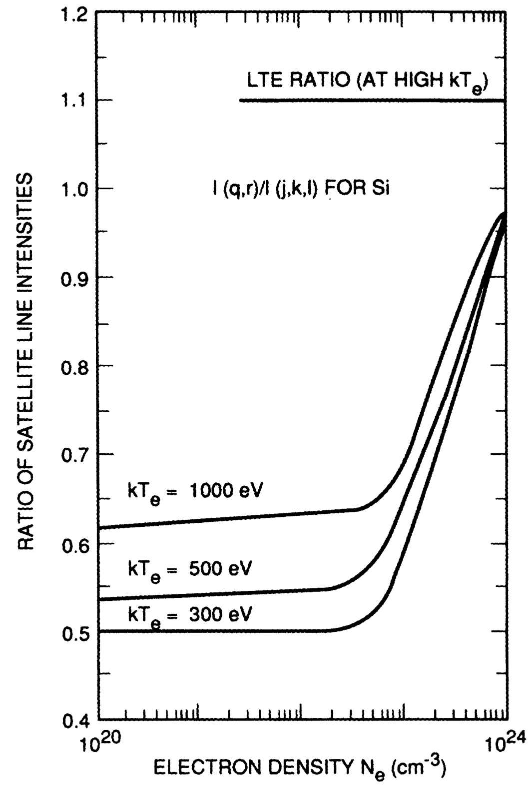

FIG. 3. The intensity ratio between the $1s2p(^1P)2s\,{}^2P \rightarrow 1s^2 2s\,{}^2S$ (q,r) satellite lines and the $1s2p^2\,{}^2D \rightarrow 1s^2 2p\,{}^2P$ (j,k,l) satellite lines for Si, as a function of the electron density N_e for various values of the electron temperature T_e.

10^{23} cm^{-3}. Since the plasma density can only decrease in time, the averaging effect of our space-time integrated spectral observations can only cause the deduced electron densities to represent a lower bound. Furthermore, since the enhanced (a,b,c,d) emission can only be produced during hot and dense plasma conditions that can exist for a very short time, the relative intensity ratio is not expected to be significantly altered by the effect of space-time integration. It should be noted that the time-integrated intensity ratio between the (a,b,c,d) dielectronic satellite lines and the (q,r) dielectronic satellite lines has been both experimentally and theoretically investigated by Burkhalter, Apruzese, and Duston [12] in a lower-density laser-produced Al plasma. The (a,b,c,d) dielectronic satellite lines observed in that investigation were relatively weak, in accord with the theoretical predictions [6].

At even higher electron densities, exceeding 10^{24} cm^{-3}, the dielectronic satellite intensity ratios can be estimated by using the Boltzmann relationship for the LTE distribution of population among the autoionizing states. At high temperatures, the LTE intensity ratios are given by

$$I(a \rightarrow f)/I(a' \rightarrow f') = (g_a/g'_a)A_r(a \rightarrow f)/A_r(a' \rightarrow f') \ , \tag{5}$$

which has been evaluated to provide the limiting values of 1.81 and 1.09 shown in Figs. 2 and 3, respectively.

IV. CONCLUSIONS

In conclusion, the analysis of high-resolution dielectronic satellite spectra of multiply charged lithiumlike ions, produced by the interaction of high-intensity, subpicosecond ultraviolet laser irradiation of solid surfaces, shows that the properties of these spectra can give a direct measure of the ambient plasma electron density in the radiating volume. It is found that the dielectronic satellite transitions originate from a plasma region with an electron density on the order of 10^{23} cm^{-3}. While it is difficult to give a precise value for the amount of the dielectronic satellite line emission that is produced at this density during such a complex laser-matter interaction process, we have argued that the relevant satellite emission must occur predominantly in a high-temperature high-density plasma, because the emitting ions are significantly abundant only under these conditions. In order to extend this investigation to even higher electron densities around 10^{24} cm^{-3}, where the plasma electric microfields together with the angular momentum changing electron-ion collisions can cause significant overlapping of the dielectronic satellite features, it would be necessary to calculate the dielectronic satellite spectra in the presence of Stark broadening, following the procedure described in a recent theoretical treatment [13]. It is the sensitivity of the dielectronic satellite spectra to a selective electron-ion collisional process, which is particularly active in the high-density region, that gives rise to the enhancement of the satellite line intensities. Finally, the experimental results have been found to be in accord with earlier theoretical work describing this radiative mechanism involving transitions from autoionizing states. This agreement between the experimental results and the theoretical predictions indicates that such satellite features can serve as a general and valuable diagnostic of high-density plasmas produced by high-intensity radiative interactions with solid matter.

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