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Experimental discovery of charge-exchange-caused dips in spectral lines from laser-produced plasmas

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We report the first experimental observation of charge-exchange-caused dips (also called *x* dips) in spectral lines of multicharged ions in laser-produced plasmas. Specifically, in the process of a laser irradiation of targets made out of aluminum carbide, we observed two *x* dips in the Ly_γ line of Al XIII perturbed by fully stripped carbon. From the practical point of view, this opens up a way to experimentally produce not-yet-available fundamental data on charge exchange between multicharged ions, virtually inaccessible by other experimental methods. From the theoretical viewpoint, the results are important because the *x* dips are the only one signature of charge exchange in profiles of spectral lines emitted by plasmas and they are the only one quasimolecular phenomenon that could be observed at relatively "low" densities of laser-produced plasmas.

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I. INTRODUCTION

Charge-exchange-caused dips (also called *x* dips) in spectral lines emitted by plasmas are a relatively new phenomenon. It was first observed in the profile of the neutral hydrogen line H_α emitted from a helium plasma of the gas-liner pinch [1]. That paper [1] also gave a sketch of the underlying theory.

Subsequent works [2,3] presented a more sophisticated theory of the *x*-dip phenomenon. These theoretical works [2,3] focused on the possibility of observing the *x* dips in spectral lines of *multicharged* ions in laser-produced plasmas. The reason for this focus is that the *x*-dips phenomenon opens up an alternative new way to experimentally obtain data on charge exchange (CE) between multicharged ions. For most pairs of multicharged ions this might be the only way to obtain such experimental information of fundamental importance. The theoretical papers [2,3] provided a list of pairs of multicharged ions, which were the most favorable candidates for observing the *x* dips in laser-produced plasmas. This list contains particular spectral lines of some hydrogenlike ions of a nuclear charge Z perturbed by some particular fully stripped ions of a nuclear charge $Z' \neq Z$.

In the present paper we report the first experimental observation of the *x* dips in spectral lines of multicharged ions in laser-produced plasmas. Specifically, in the process of a laser irradiation of targets made out of aluminum carbide, we observed two *x* dips in the Ly_γ line of Al XIII perturbed by fully stripped carbon.

II. GIST OF THE *X*-DIP PHENOMENON

A comprehensive theory of the *x*-dip phenomenon was presented in [2,3]. Here we give only some brief excerpts from [2,3].

We consider electron terms in the field of two stationary Coulomb centers (TCC's) of charges Z and Z' separated by a distance R. Gershtein and Krivchenkov [4] demonstrated that the well-known Neumann-Wigner general theorem on the impossibility of crossing of terms of the same symmetry [5] is not valid for the TCC problem of $Z' \neq Z$. Physically it is a consequence of the fact that the TCC problem allows a separation of variables in the elliptic coordinates [4].

The overwhelming majority of crossings in the case of $Z' \neq Z$ are real, not avoided crossings. Only a tiny minority are avoided crossings. An avoided crossing occurs when two wells, corresponding to separated Z and Z' centers, have states Ψ and Ψ', characterized by the same energies $E = E'$, by the same magnetic quantum numbers $m = m'$, and by the same radial elliptical quantum numbers $k = k'$ [6–8]. In this situation, the electron has a much larger probability of tunneling from one well to the other (i.e., of CE) as compared to the absence of such degeneracy. Physically, this selection rule follows from the picture of CE as the corresponding interaction of states in two adjacent potential wells (one centered at the charge Z, another at the charge Z') and from the fact that for such interaction to be possible, the radial wave functions of these states should have the same number of nodes [7,8].

We consider a radiative transition between two terms corresponding to some Stark component. We use atomic units and therefore employ the same notation $f(R)$ for both the transition energy and the transition frequency; R is the distance between the radiator and the perturbing atom or ion. We focus at a vicinity δR of some particular distance R_0 corresponding to a small part $\delta\omega$ of the component profile around $\Delta\omega_0 = f(R_0)$.

Let us consider a term a of a principal quantum number n, which asymptotically (at $R \to \infty$) is a radiator's term, and a term a' of a principal quantum number n', which asymptotically is a perturber's term. We are interested in the situation where in the vicinity of R_0 there occurs an avoided crossing of the term a with the term a'. It translates into an avoided crossing in the energy difference between upper terms a, a' and a lower term a_0 (see Fig. 1).

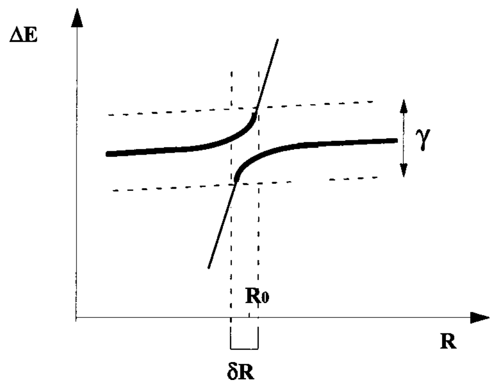

FIG. 1. Transition energies $f_{-}(R)=E_a-E_{a0}$ and $f_{+}(R)=E_a-E_{a0}$ versus the radiator-perturber separation R, plotted in a vicinity δR of an avoided crossing of the perturber's term a' with the radiator's term a at $R=R_0$ in the course of the radiative transition from the term a to the term a_0. The plot is schematic. The transition energy $f_{-}(R)$ actually occupies a band of a width γ (shown by dashed lines) controlled primarily by the dynamical broadening caused by electron and ion microfields in a plasma. The radiator's transition energy modified by the avoided crossing is shown by the bold line. In the interval δR, the transition energy has two branches, corresponding to the fact that the wave function of the radiator's term in this interval is a linear combination of wave functions of two different energies.

For the majority of radiators the perturbing ion of the charge Z' is separated by a distance R that is not in a close proximity to R_0. In this case the formation of the line profile in the line wings can be described in the usual way by the splitting of the spectral line into Stark components due to a static electric field $F=Z'/R^2$ produced by the charge Z'. The Stark components are broadened by electron collisions and coupled by nondiagonal terms of the electron broadening operator [1]. The observed profile is the superposition of the collisional contours emitted at different values of F, that is, at different values of R.

The situation differs dramatically for a minority of radiators for which the distance R is close to R_0 [1]. Because of the avoided crossing of the terms, CE appears as an *additional channel for decay* of the excited state of the Z ion. Therefore the lifetime of the excited state is shorted and the collisional width *suddenly* increases. This means that these radiators emit a broader collisional profile (compared to the others), which is consequently less intense in its central part and more intense in its wings. Thus at the frequency $\Delta\omega[F(R_0)]$ the intensity of the resultant line profile becomes smaller than it would be if the avoided crossing was absent. As a result, a dip might appear in the line profile.

In the comprehensive quantitative theory developed in [2,3] it was shown, in particular, that when a CE-caused avoided crossing occurs at a relatively large distance $R \gg \max(n^2/Z,n'^2/Z')$, then it *practically always results in a dip* in the profile of the corresponding Stark component of the spectral line.

In [2,3] there were also quantitatively determined upper and lower limits that control the range of electron densities, where the x dips can be observed. Both the lower limit and one out of two competing values of the upper limit physically come from the requirement that the crossing distance R_0 should not differ too much from the most probable internuclear distance. Another upper limit physically comes from the condition that the dynamical Stark broadening by electrons and ions of the plasma should not be so large as to wash out the dip.

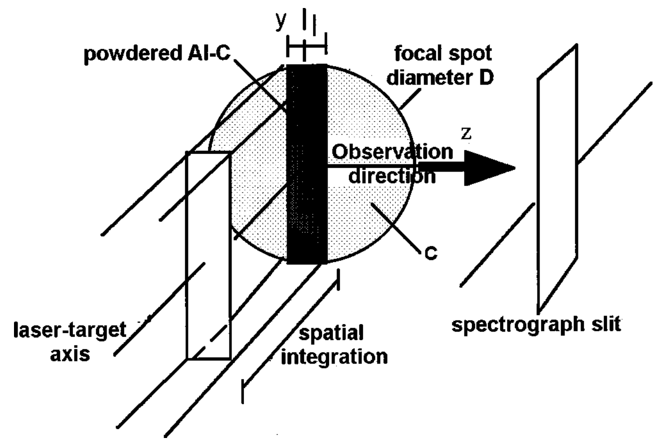

FIG. 2. Experimental setup, including the design of the structured target. The aluminum carbide plasma is confined in a carbon plasma in the direction of the observation, thus allowing the control of the reabsorption. The Al_4C_3 strips, centered on the laser beam, are well suited for the optimization of the emission. The spectrograph slit ensures a spatial resolution along the laser-target axis.

This theory was applied in [2,3] for selecting prospective ZeZ'-systems for observations of x dips in laser-produced high-density plasmas of multicharged ions. One of the prospective candidates was the Ly_γ line of Al XIII ($Z=13$) perturbed by fully-stripped C ($Z'=6$). Avoided crossing of some of the $Z=13$ terms of $n=4$ with some of the $Z'=6$ terms of $n'=2$ occur in a vicinity of $R_0\approx 8$. This distance is about 6.5 times greater than the size n^2/Z of the largest out of the two separate states (Z,n) and (Z',n'). More specifically, two out of these avoided crossings should translate into x dips located at $\Delta\lambda\approx 6.7$ and $\Delta\lambda\approx 9.6$ mÅ (i.e., on the red side). The range of densities where these two x dips could be observed (for $T\sim 400$ eV) is $N_e^{\mathrm{upper}}\approx 1\times 10^{22}$ cm^{-3}, $N_e^{\mathrm{lower}}\approx 2\times 10^{20}$ cm^{-3}.

III. EXPERIMENT

The experiment has been performed at the nanosecond laser facility at the LULI, France. When a high intensity (4×10^{14} W/cm^2) 4ω laser beam in a pulse of 500 ps is focused onto a massive target, the electron density and temperature can reach extreme values such as 10^{23} cm^{-3} and 300 eV, respectively, in the first 5 μ of the plasma inside the crater. The density and the temperature gradients in the adjacent layer of 10–15 μ can be easily diagnosed by emission spectroscopy. The experimental setup designed to optimize both the generation and the diagnostics of the x dips in the Ly_γ line of Al XIII perturbed by fully stripped carbon is shown in Fig. 2.

The targets are structured; powdered aluminum carbide (Al_4C_3) strips are inserted in a carbon substrate. For each

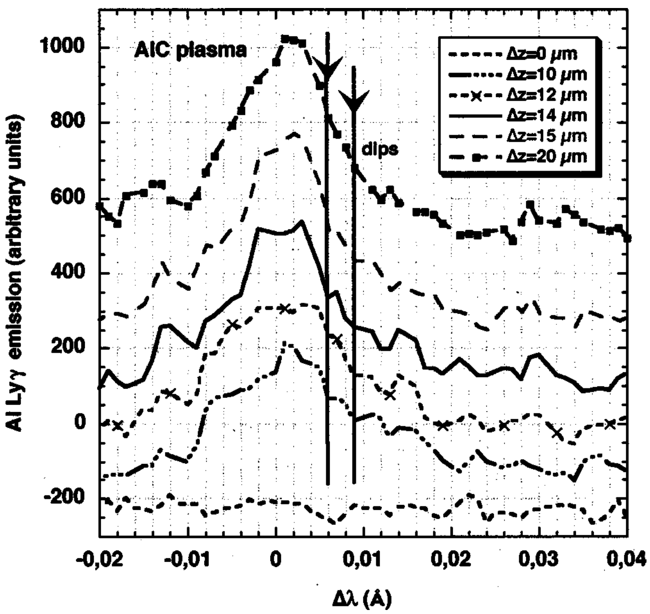

FIG. 3. Experimental profiles of the Ly_γ line of Al XIII emitted from an aluminum carbide plasma. There is shown the evolution of the spectra as the spatially integrated slice Δx increases in thickness from $\Delta x = 0$ at the bottom of the crater, until it includes the corona plasma ($\Delta x = 20\ \mu$m). For $5 \leqslant \Delta x \leqslant 15\ \mu$m, corresponding to the densities 10^{20}–10^{22} cm^{-3}, the profiles exhibit two pronounced dips in the red wing. For $\Delta x \geqslant 20\ \mu$m, only discontinuities of the slope of the line profile in the red wing are still visible as remnants of the *x* dips. In the latter case, the average density for the selected plasma slice is getting too low for observing the *x* dips, the densest part of the plasma being hidden by the slit (15 μm).

target, the Al_4C_3 strip is placed through the center of the focal spot ($\varnothing = 80\ \mu$m). They are well suited for the optimization of the emission intensity and for the control of both the transverse inhomogeneities and the reabsorption. It turns out that, for the density domain 10^{20}–10^{22} cm^{-3} required for observing the *x* dips in the Ly_γ line of Al XIII, thin strips of the thickness 20 μm serve well the above purpose [9].

A diagnostic of an ultrahigh spatial and spectral resolution records spectra emerging from progressively thicker slices of plasma, perpendicularly to the laser beam, with the help of the 15-μm slit. This slit is located at 2 mm from the plasma so that the high transverse magnification of about 100 facilitates the analysis of the inherent spatial gradients. As for the spectral resolution ($R = 8000$), it is achieved by using a PET crystal set up in the Johann geometry and employed at the first order.

Figure 3 shows the evolution of the experimental profile of the Ly_γ line of Al XIII as the spatially integrated slice Δx increases in thickness from $\Delta x = 0$ at the bottom of the crater until including the corona plasma ($\Delta x = 20\ \mu$m). For all the laser shots corresponding to the same conditions, the observed spectra demonstrate the same qualitative features.

Because the spectrograph is inclined, the emission from the entire dense shock-region plasma is accessible, corresponding to the first spatially integrated slice of $\Delta x = 5\ \mu$m. Between 5 and 15 μm, the spatial integration involves a progressively less dense plasma and every profile exhibits two pronounced dips in the red wing. The dips are located at 6 and 9 mÅ from the center of the line. These positions are close to the predicted positions [3] (6.7 mÅ and 9.6 Å, respectively), which had been calculated analytically using the assumption (5).

It has been checked by hydrosimulations [10] that the electron densities involved in the spatial integration interval 5–15 μm are consistent with those optimizing the visibility of the dips in the wing. For a larger thickness of the spatial integration, i.e., $\Delta x \geqslant 20\ \mu$m, only discontinuities of the slope of the line profile in the red wing are still visible as remnants of the *x* dips. In the latter case, the average density for the selected plasma slice is getting too low for observing the *x* dips.

IV. CONCLUSIONS

We presented an experimental discovery of the *x*-dip phenomenon in spectral lines of multicharged ions in laser-produced plasmas. This phenomenon of a *multidisciplinary* character (bringing together atomic physics and plasma physics) should have a significant practical importance. Indeed, further experimental studies of *x* dips, using various ''radiator-perturber'' pairs, would serve for producing not-yet-available fundamental data on CE between multicharged ions, virtually inaccessible by other experimental methods.

The *x*-dip phenomenon is also important from the theoretical point of view for the following reasons. Let us compare this phenomenon to quasimolecular satellites (QS) observed in spectral lines emitted by laser-produced plasmas [9,11–14]. The QS result from extrema in transition energies *unrelated to avoided crossings*. The *x* dips and the QS have one thing in common: in the course of a collision, for a relatively short period of time, the electron is shared by two ionic centers—a transient molecule (quasimolecule) is formed. However, other features of the *x*-dip phenomenon are unique and clearly distinguish it from the QS.

First, for a given pair of multicharged ions, the *x* dips are observed in plasmas of densities by two or three orders of magnitude lower than densities, at which the QS are observed. This is because the internuclear distance, corresponding to avoided crossings responsible for the *x* dips, is typically an order of magnitude greater than the internuclear distance, corresponding to extrema in transition energies unrelated to avoided crossings. Coupled with the requirement that, for favorable observation conditions, the mean interionic distance in a plasma should be close to the corresponding internuclear distance of interest, this explains the above dramatic difference in favorable plasma densities. Thus, the *x* dips are the only one quasimolecular phenomenon that could be observed at relatively ''low'' densities of laser-produced plasmas.

Second, it should be emphasized that—distinct from the *x* dips—the QS can be unrelated to CE (despite statements to the contrary in some papers). The mere fact that, for a relatively short time, the electron is shared between two ionic centers does *not* necessarily mean that CE would occur. Indeed, CE requires that the electron, which was initially bound at one ionic center, would finally end up at the other ionic center.

In this paper we focus at pairs consisting of two different ionic centers, since only such pairs are relevant to the *x* dip phenomenon. For pairs consisting of two different ionic centers, CE does *not* happen in the process leading to the QS for the following reason. CE would require a coupling of those two quasimolecular terms that asymptotically (at $R \to \infty$) would correspond to the electron being bound by only one or the other ionic center. If the coupling was there, then at relatively small R, where these two terms come close together, an avoided crossing would occur. However, in reality, at the internuclear distance corresponding to an extremum in transition energies, there is no avoided crossing of the terms responsible for the QS and therefore no CE occurs.

Thus, the bottom line is that the *x*-dip phenomenon is *the only one signature of CE* between nonidentical ionic centers in profiles of spectral lines emitted by plasmas. This further enhances the importance of our first experimental observation of the *x*-dip phenomenon in laser-produced plasmas. Our experimental study of this phenomenon in laser-produced plasmas containing other ''radiator-perturber'' pairs is ongoing. A preliminary analysis of the experimental spectra is encouraging; results on the *x* dips involving other radiator-perturber pairs are expected to be presented elsewhere in the near future.

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