Dynamics of Collisional-Excitation X-Ray Lasers

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We present absolutely timed spectral measurements, as well as other data, on the exploding-foil Ne-like Se soft-x-ray laser scheme. The data show that amplification occurs near the peak of the optical heating pulse, in agreement with our original theoretical predictions. A recently published alternative scenario predicted that gain develops only after the heating pulse, via recombination in an overionized, rapidly cooling plasma. Since our new data differ so sharply from that prediction, we reexamine the theoretical basis for that alternative scenario.

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In the summer of 1984, a series of experiments, using a high-power optical laser to heat and explode thin foils[1] of Se succeeded in demonstrating[2] amplification of Ne-like $3p$-$3s$ transitions at 206 and 209 Å. Whereas higher gains were expected[1] at 182 Å (a $J=0$ to 1 transition), that line could not even be uniquely identified[2] in the presence of nearby lines (for short gain lengths), while the aforementioned two $J=2$ to 1 lines demonstrated gains of 5 cm^{-1} roughly in accord with theory. While recent experiments[3] (under identical conditions but at longer lengths which allowed us unambiguously to observe gain) showed amplification of the 182-Å line, the gains of about $\frac{1}{2}$ those of the $J=2$ lines are still quite low compared with theory. Recently an explanation for the $J=0$ anomaly and $J=2$ success was offered in a paper[4] to which we shall refer as ADBKJ, after the authors' initials.

The ADBKJ explanation can be summarized as follows: While the driving laser is on, the 1-keV plasma is stripped well past the Ne-like ionization state. The key controlling feature here is a dielectronic recombination rate coefficient of 3×10^{-12} cm^3 s^{-1}, nearly an order of magnitude lower than that of Ref. 1. With virtually no Ne-like ions present when the plasma is hot, there is no gain on any Ne-like line, in particular, the $J=0$ to 1 (in contradiction to the recent measurements). In addition, these conditions should lead to high gains on F-like lines,[5] yet to date they have been too small to be measured. To explain the observed $J=2$ to 1 gain, ADBKJ invoke rapid (on a 100-ps time scale) radiative cooling when the driving laser turns off, assuming a minimum radiative rate of 2.4×10^{-26} W cm^3 (per ion per electron), and an electron density n_e of 10^{21} cm^{-3} at that time. As the Se recombines to the Ne-like state, the $J=2$ levels are preferentially populated over $J=0$, and the plasma is now too cool for collisional excitation of the $J=0$. They then predict a gain of 5 cm^{-1} for the $J=2$ to 1 transitions in this late-time regime, assuming $n_e=5\times10^{20}$ cm^{-3}. In a variation on this ADBKJ "late-time" theme, Hagelstein[5] has speculated that strong scattering while the laser pulse is on would prevent beam propagation and lead to no observed gain on any line. Late in time with the propagation restored after the pulse is off, $J=2$ gain might be observed, but not F-like gain and possibly not $J=0$ gain.

While we offer no definitive explanation for the low gain observed on $J=0$, our views of the dynamics of the situation are quite different from those of ADBKJ. We believe that there is a significant fraction of the Se in the Ne-like state during optical laser pumping ($\simeq20\%$) and that the $J=2$ laser is pumped in a quasicontinuous manner, partially by collisional excitation and partially by dielectronic and three-body recombination from F-like states and by cascade from higher Ne-like states, as stated, in part, in the second sentence of Ref. 1.

Several experiments can differentiate between these two hypotheses. The clearest is a measurement of the timing of the x-ray signal, which can show whether gain is occurring while the optical laser pump is on[1] or after it turns off as predicted by ADBKJ. Secondly, its duration should scale to some degree with laser pump duration according to our scenario. Finally, spectral evidence for dominant Ne-like emission early in the optical laser pulse, not late, should also support our view of significant Ne-like fractions while the plasma is hot. In what follows we present new data which indeed support our picture of the dynamics in the ways just described. Later we reexamine the theory upon which ADBKJ's alternative scenario is based.

The absolute-time history of the Se x-ray laser lines was measured with use of a transmission-grating streaked spectrometer (TGSS).[6] The time reference was provided by an optical fiducial pulse of known timing relative to the peak of the Nova-laser drive pulse.[7] Measurements were made along two different lines of sight, along the foil axis as well as 7 mrad off axis, in order to account for the dynamics of the amplified x-ray beam propagation. Early in time, the density gradients are steep and the beam is refracted off of the foil axis. Later in time, the gradients relax and the beam can propagate down the laser axis.[8,9] The acceptance angle for the TGSS is 7 mrad. Figure 1 displays the time history of the $J=2$ to 1 206-Å line for both the off-axis and on-axis measurements. (Target and irradiation conditions were identical to those of Ref. 2, except that this result is the output of a 3-cm-long target.) The optical driving laser is also shown. The on-axis x-ray laser emission peaks near the peak of the drive laser. Moreover, the off-axis x-ray laser emission peaks even $\simeq 100$ ps earlier, *before* the peak of the drive laser. The absolute-timing uncertainty is ± 120 ps. The dashed curve that nearly overlies curve b is our prediction for the on-axis lasing signal. It is the result of the convolving of predicted gain and density-profile time histories by the methods of Ref. 1, with a calculation of the effects of refraction on beam amplification and propagation down the axis. The

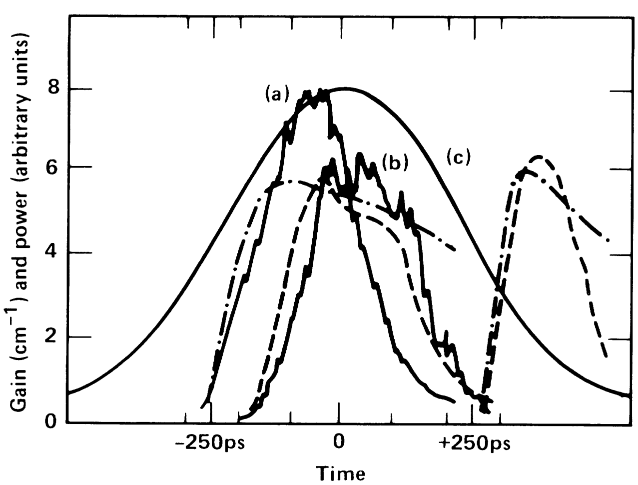

FIG. 1. Power (arbitrary units) vs time of 206-Å lasing line (curve a) off axis and (curve b) on axis compared with driving laser pulse (curve c). Dot-dashed curve under c is predicted gain vs time from model of Ref. 1, and dashed curve is the predicted on-axis laser emission from that model, which compares quite well to the corresponding data curve b. The dot-dashed and dashed pair to the far right (after the back half of the drive curve c) are our estimates of the corresponding gain and emission curves from the model of Ref. 3.

dashed curve to the far right is our best guess for the equivalent prediction of ADBKJ and is clearly ruled out by the data. This also rules out Hagelstein's speculation,[5] as he points out himself in his note added in proof.

Independent confirmation of these timing data comes from absolutely timed beam divergence data,[9] obtained with coarse spectral resolution via an L-edge filter/mirror combination. Those on- and off-axis data, absolutely timed to ± 70 ps, and obtained on a separate detector, agree with the TGSS data presented here, and deepen our confidence in these results.

For a 1-ns 0.53-μm laser pulse (twice the normal duration) on an appropriately scaled 1500-Å-thick-Se/1500-Å-Formvar target, irradiated at 2×10^{13} W/cm^2 per side, we have observed in excess of 50% longer temporal history of the lasing lines, supporting our quasicontinuous view of the lasing process. (In this long-pulse experiment, the $J=0$ laser line still has relatively low gain.)

A possibly misleading clue to the dynamics lies in Fig. 3(b) of Ref. 2, which shows nonabsolutely timed evolution of a $J=2$ laser line emission as well as a Na-like resonance-line emission (sighted down the axis of a 1-cm target so that they are both within the dynamic range of the instrument). The figure shows both lines rising at about the same time, which could be interpreted as late-time lasing during the recombining cooling phase when the Na-like fraction builds up, as per ADBKJ. However, our calculations show both lines rising *during* the optical laser pulse, not later. The optically thick (mean free path $\simeq \frac{1}{2}$ mm) Na line emission increases simply because the source area increases as the foil expands.

The time-resolved $3d$-$2p$ and $3s$-$2p$ x-ray spectra from Ne-like and F-like states presented in Ref. 1 were the original motivation for our belief that significant amounts of Ne-like Se ions were present during the driving laser pulse. While the analysis of these spectra was complicated by issues of optical depth, and by the existence of cooler, nondirectly illuminated regions surrounding the driving laser line-focal spot, to date our best 2D simulations with line transfer, using the methods described in Ref. 1, support the view that the signal comes from the central lasing region. As a check, we have performed an experiment with a 100-μm-high strip of Se (750 Å thick, on a 1500-Å CH foil as usual) illuminated by a 200-μm-high line focus which overfills the strip. This target performed quite similarly to a standard "full" Se foil both in its 3-3 lasing and in its 3-2 spectral time history. In particular, these time-resolved, spatially localized data show prominent Ne-like line emission. Our modeling indicates that the major part of this signal occurs during the driving pulse when the foil is relatively dense, and not after it turns off when the density is so much lower. Thus a significant fraction of the hot plasma is in the Ne-like state.

Thus, both from the extreme ultraviolet (XUV) laser-signal absolute timing and from the 3-2 x-ray spectra, it is clear that while the driving laser is on and the plasma is hot, there is a significant fraction of Ne-like ions, and gain is occurring then, not later as per ADBKJ. We therefore must reexamine the theory that formed the basis for the ADBKJ scenario.

The key element in ADBKJ's prediction of nearly no Ne-like Se during the driving laser pulse was the dielectronic recombination rate coefficient of $3\times10^{-12}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$ (from F-like to Ne-like). The references quoted for this value were attributed to one of us (V.L.J.). Those calculations were based on an inappropriate extrapolation to Se of a theory valid only for low-Z elements. For low-Z elements, dielectronic recombination is reduced by the Coster-Kronig processes ($\Delta n_c = 0$ radiationless transitions of the excited core electron accompanied by the return of the captured nl electron to the continuum). For Ne-like Se, the dominant contributions to the dielectronic recombination rate arise from electron capture into the $n=3$, 4, and 5 levels, for which the $\Delta n_c = 0$ ($n_c = 3$ to 3) Coster-Kronig processes are not energetically permissible. Thus, low-Z models, in which the Se $\Delta n_c = 0$ processes are assumed to occur for all outer-electron n values, will be invalid for Se and will underestimate the dielectronic recombination rate.[10] When this disallowed Coster-Kronig process is removed from the calculation, recombination coefficients 10 times higher [of order $(2\text{–}3)\times10^{-11}\ \mathrm{cm}^3\ \mathrm{s}^{-1}$] are found,[11] in reasonable agreement with recently reported calculations.[12,13] With the higher, more realistic recombination rate, about 20% of the 1-keV plasma should be Ne-like, permitting $J=2$ to 1 lasing during the driving laser pulse as observed. Since under these (hot, 20% Ne-like) conditions even ADBKJ would predict significant gain, the weak $J=0$ to 1 gain reappears as a mystery to be explained.

We turn now to the issue of radiative cooling. We find the basic radiative rate of $2.4\times10^{-26}\ \mathrm{W\ cm}^3$ at $n_e = 10^{21}\ \mathrm{cm}^{-3}$, that ADBKJ quote, to be quite reasonable. Our best model,[3,11] including dielectronic recombination and $\Delta n = 0$ transitions, predicts 2.8×10^{-26} W cm^3. At very low densities ($6\times10^{13}\ \mathrm{cm}^{-3}$) this model predicts rates nearly 3 times higher in agreement with an interpolated value (between Fe and Mo) for Se of Jensen *et al.*[14] This density dependence is not due to collisional deexcitation as a competitor to radiation but rather to a change in ionization balance. At $n_e = 10^{21}\ \mathrm{cm}^{-3}$ we find the time to cool from 1 to 0.2 keV to be over 300 ps not the 130 ps quoted by ADBKJ. In arriving at our answer, physical effects of importance are the calculated change of 2.5 in charge state and the near-linear drop in radiative rate as T_e drops.

Far more critical, however, is the ADBKJ assumption that $n_e = 10^{21}\ \mathrm{cm}^{-3}$ when the driving laser turns off in order to allow radiative cooling. Our hydrodynamic simulations,[1] as well as similarity solutions that include hydrodynamics as well as laser heating,[15] show that n_e is somewhere between (1 and 2)$\times10^{20}\ \mathrm{cm}^{-3}$ by the time the laser falls to half its peak power. Thus, since the radiative cooling is roughly proportional to density, it is 5 to 10 times slower than at $10^{21}\ \mathrm{cm}^{-3}$ as assumed by ADBKJ. Thus it is no surprise that the observed timing of the $J=2$ lasing contradicts the prediction of rapid radiative cooling (and late-time lasing), which was not based on a self-consistent hydrodynamic calculation and thus used a density (and hence a cooling rate) nearly an order of magnitude too high.

Holographic interferometry was performed at KMS Fusion, Inc., with parameters nearly identical to our nominal gain experiments: two-sided illumination, multiple frames, $4.5\times10^{13}\ \mathrm{W/cm}^2$ per side, 0.53 μm, 470-ps-wide flat-top pulse, and 750-Å-thick Se. Our simulations indicate that the flat top (versus Gaussian) and round spot (versus line focus) do not significantly change the density-profile results. The data agree closely with our simulations, and demonstrate directly that $n_e \simeq 10^{20}\ \mathrm{cm}^{-3}$ at the end of the laser pulse.[16] The holography results of Fig. 2 in Ref. 1, which show somewhat higher densities, were the best available at the time. Its clearly described conditions (shorter pulse, one-sided illumination, and lower irradiation) all conspire to raise the density above that of the nominal case, and was meant to show our ability correctly to model and predict $n_e(x,t)$. Inferences of the density evolution derived from time-resolved spectra of Raman back-scattered optical laser light[17] are in close agreement with the calculations as well (about $3\times10^{20}\ \mathrm{cm}^{-3}$ at the laser peak and $2\times10^{20}\ \mathrm{cm}^{-3}$ about 200 ps later). Moreover, inferences of T_e from those data reveal an upper limit to the temperature of about 1.3 keV at the peak of the laser drive, dropping to about 700 eV 250 ps later, consistent with the simulations. This is not to say that radiative cooling cannot ever be important. Indeed we concur with ADBKJ that targets could and should be designed to exploit radiative cooling as a way to enhance gain for short-wavelength, recombination-pumped x-ray lasers.

A final point worth examining is their gain calculation for the $J=2$ lasing, even assuming an inordinately rapid radiative cooling rate. The gain of 5 cm^{-1} is based on an n_e of $5\times10^{20}\ \mathrm{cm}^{-3}$. Since at this late time n_e is closer to $1\times10^{20}\ \mathrm{cm}^{-3}$, to first order, the actual "calculated" gain at this time should be 1 cm^{-1}, in rather poor agreement with the data. Thus, when we take into account the proper n_e (based on self-consistent hydrodynamic calculations and supported by data), the ADBKJ scenario of 100-ps time-scale rapid radiative cooling followed by recombination-pumped gain of 5 cm^{-1} fails to hold together. More importantly, coupled with our earlier discussion of dielectronic recombination rates and clear time-resolved data showing reasonable Ne-like fractions while the plasma is hot, the low $J=0$ gain cannot be accounted for either. A variation on ADBKJ, in which the hot overstripped plasma is separated from the cool recombining one not in time, but in space, is not supported quantitatively by our current calculations. Nor would it explain the lack of F-like lasing lines. However, experiments are planned to pursue this hypothesis by better localization of the lasing regions.

Some recent data bear on these issues. Observations of amplification[18] on Ne-like Mo include a measured gain of about 2 cm^{-1} for the $(\frac{3}{2},\frac{3}{2})_0 \rightarrow (\frac{3}{2},\frac{1}{2})_1$ $3p$-$3s$ transition at 106 Å. This is within a factor of 2 of theoretical expectations[18] for this collisionally excited laser. [The equivalent transition in Ne-like Se at 169 Å was predicted to have low gain (1 cm^{-1}) compared with the $(\frac{1}{2},\frac{1}{2})_0 \rightarrow (\frac{1}{2},\frac{1}{2})_1$ 182-Å line as a result of its relatively low collisional excitation rate.] Nonetheless, the gain of the 182-Å $J=0$ to 1 Se equivalent in Mo at 141 Å, also predicted to be of order 3 cm^{-1}, was too small to be measured. In retrospect, then, any theory that wishes to account for the low Se $J=0$, on the basis of purely hydrodynamic variables, must also account simultaneously in Ne-like Mo for a $J=0$ to 1 line that shows close agreement with theory and one that does not. Alternative scenarios such as Griem's,[19] which invoke very selective line-broadening effects (coupled with turbulence), may be able to account for the Mo result as well as the Se. Another possibility is selective absorption,[5] possibly from Mg-like or Al-like states, of the missing laser lines. These scenarios may be difficult to prove or disprove experimentally but must be pursued if we are to make further progress in accounting for the weak gain on the $J=0$ line. Recent gain studies on lower-Z systems may also shed light on these issues.[20]

In summary, absolutely timed spectral measurements show that gain occurs near the peak of the heating pulse in agreement with our standard model and in distinct disagreement with the alternative scenario of ADBKJ. We believe that more accurate treatments of dielectronic recombination and the electron-density evolution as presented here can account for the major differences between the two models. The quantitative behavior of the 182-Å $J=0$ to 1 line (now only weak, rather than missing), absent F-like gain, and the Mo results are yet to be explained.

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