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Collisional Redistribution Effects On X-ray Laser Saturation Behavior

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Abstract. We recently published a detailed summary of our experimental and theoretical research on Ne-like Se x-ray laser line widths, and one of our conclusions was that collisional redistribution rates are likely to have an effect on the saturation behavior of the 206.4 Å Se x-ray laser. In this paper we focus on the effects of collisional redistribution on x-ray laser gain coefficients, and discuss ways of including these effects in existing laser line-transfer models.

Over the past several years, we have obtained high-resolution spectra of several Ne-like x-ray laser transitions, including the $(3/2,3/2)J=2 - (3/2,1/2)J=1$ lasers in $Y^{1,2}$ (155.0 Å) and $Se^{3,9}$ (206.4 Å) and the $(1/2,1/2)J=0 - (1/2,1/2)J=1$ lasers in Se^9 (182.4 Å), Nb^{10} (145.9 Å) and Zr^{10} (150.4 Å), using a unique high-resolution soft x-ray spectrometer^{11,9}. These experiments showed gain-narrowing of the 206.4 Å and 182.4 Å Se lasers with increasing amplifier length, showed no saturation rebroadening of the 206.4 Å Se laser in long, saturated amplifiers, and demonstrated hyperfine line splitting in the soft x-ray regime. It is now possible to measure soft x-ray line profiles with unprecedented resolution, and we have used this technique in other experiments to observe time-dependent Stark broadening in an expanding, rarefying plasma¹² and to measure relative wavelength separations with high precision².

We recently published a detailed summary of our experimental and theoretical research on Ne-like Se x-ray laser line widths and their variation with amplifier length⁹. In that research, we measured the line width of the 206.4 Å Se laser from amplifiers of varying lengths, and compared the resulting data to laser line profile and line transfer calculations based on the expected temperature and density of the Se plasma. In particular, we found that the observed lack of inhomogeneous saturation rebroadening of this laser in long, saturated amplifiers could be attributed to: 1) a rapid decrease in saturation rebroadening induced by even relatively small homogeneous lifetime broadening contributions to the intrinsic line profile, and 2) collisional redistribution effects which would be expected to effectively homogenize the Doppler component of the intrinsic line profile. We refer the reader to that paper⁹ for the details of the experiments, the calculations and the conclusions, which cannot be reproduced in this short proceedings paper.

Instead, in this paper we discuss in more detail the effects of collisional redistribution on laser gain coefficients, and discuss ways of including these effects in existing amplified spontaneous emission (A.S.E.) laser line transfer models. Again, we refer the reader elsewhere^{8,9} for details which we must omit from the present paper due to space constraints.

We begin with a simple three-level collisional excitation model for Ne-like x-ray lasers, where inversion is attained and maintained through collisional excitation of the upper laser level from the ground state, and all other levels are ignored. We will initially ignore collisional redistribution effects, and will derive an expression for the frequency-dependent gain coefficient in their absence.

The velocity-dependent rate equations for a three-level laser can be written as^{13,14}

$$\frac{dN_1(v,t)}{dt} = -N_1(v,t)(\Gamma_{12} + \Gamma_{13}) + N_2(v,t)(\Gamma_{21} + A_{21}) + N_3(v,t)\Gamma_{31} \quad (1)$$

$$\begin{aligned} \frac{dN_2(v,t)}{dt} = & N_1(v,t)\Gamma_{12} - N_2(v,t)(\Gamma_{21} + \Gamma_{23} + A_{21} + R_{23}(v)) \\ & + N_3(v,t)(\Gamma_{32} + A_{32} + R_{32}(v)) \end{aligned} \quad (2)$$

$$\begin{aligned} \frac{dN_3(v,t)}{dt} = & N_1(v,t)\Gamma_{13} + N_2(v,t)(\Gamma_{23} + R_{23}(v)) \\ & - N_3(v,t)(\Gamma_{31} + \Gamma_{32} + A_{32} + R_{32}(v)) \end{aligned} \quad (3)$$

The populations $N_{1,2,3}(v)$ are the population densities of atoms in the ground state, the lower laser level and the upper laser level, respectively, per unit velocity with axial velocity v , and R_{32} and R_{23} are the stimulated emission and absorption rates. The other terms in the equations are the spontaneous emission and electron collisional excitation/de-excitation rates between the levels (A 's and Γ 's above; the spontaneous 3 - 1 transition is dipole-forbidden in the Se laser). Rates to and from other levels are neglected in order to maintain a closed system, with $N_1 + N_2 + N_3 = N = \text{constant}$.

The above equations can be solved analytically in steady-state, and the resulting frequency-dependent laser gain coefficient can be written as⁹

$$g(v) = \int \sigma_{32}(v, v)(N_3(v) - \frac{g_3}{g_2} N_2(v)) dv \quad (4)$$

$$= \frac{g_0}{V(v_0)} \int_0^\infty \frac{S(u)\phi(v, u) du}{1 + \frac{1}{I_{sat}} \int_0^\infty I(v')\phi(v', u) dv'} \quad (5)$$

where $S(u)$ is the area-normalized Doppler line profile in inverse-frequency units, $\phi(v, u)$ is the area-normalized homogeneous line profile with Doppler-shifted central frequency u , g_0 is the small-signal gain coefficient at line center, I_{sat} is the saturation spectral intensity and $V(v_0)$ is the intrinsic line profile function (a Voigt function, the convolution of $\phi(v, u)$ and $S(u)$) evaluated at line center. $I(v)$ is the laser spectral intensity in $\text{W cm}^{-2} \text{ Hz}^{-1}$.

The above rate equations are inhomogeneous in the sense that there are no rates transferring population between velocity groups, so that a single velocity is associated with each radiator. This approximation is reasonable when the velocity-

changing collision rate (the collisional redistribution rate) is large compared to the inverse of the effective lifetime of the radiator (i.e. the homogeneous line width in frequency units), so that Doppler broadening can be considered to be an inhomogeneous broadening mechanism. However, when this is not the case, essentially different saturation behavior can be expected in A.S.E. lasers, as has been pointed out before⁶⁻⁹ and was independently noted in a recent paper¹⁵. The velocity-changing collision rate is the same rate relevant to Dicke line narrowing^{16,17}, which occurs essentially when this rate Γ is larger than the Doppler width in frequency units. The condition for collisional redistribution effects is less stringent, however, since in the present case the Doppler width of the Se laser (36 mÅ) is several times larger than the homogeneous lifetime-broadening width (14 mÅ), and recent molecular dynamics simulations¹⁸ furthermore suggest that the effective collisional redistribution rate is larger than the lifetime-broadening width but smaller than the Doppler width, so that collisional redistribution effects likely are present but Dicke narrowing effects likely are not.

The essential modification to the above rate equations in order to include collisional redistribution effects is the addition of two terms to each of eqs. (1), (2) and (3) which move population into and out of a particular velocity group^{19,20},

$$\frac{dN(v,t)}{dt} \rightarrow \frac{dN(v,t)}{dt} + \int N(v',t)P(v' \rightarrow v)dv' - N(v,t) \int P(v \rightarrow v')dv' \quad (6)$$

where $P(v \rightarrow v')$ is the rate (per unit velocity) at which collisions change a radiator velocity from v to v' . The detailed form of these terms depends on the details of the collisions, and in general gas kinetic-type equations (e.g. Fokker-Plank) should be used²¹, but the strong collision limit^{19,22} provides a fairly simple means of illustrating the effect. In this limit, the velocity of a radiator after a collision is independent of its velocity prior to a collision, and the above terms are simply related to the collisional re-distribution rate Γ and the velocity distribution function $f(v)$,

$$\begin{aligned} P(v \rightarrow v') &= \Gamma f(v') \\ P(v' \rightarrow v) &= \Gamma f(v) \end{aligned} \quad (7)$$

so that for example eq. (3) becomes, in steady-state,

$$\begin{aligned} 0 = N_1(v)\Gamma_{13} + N_2(v)(\Gamma_{23} + R_{23}(v)) - N_3(v)(\Gamma_{31} + \Gamma_{32} + A_{32} + R_{32}(v) + \Gamma) \\ + \Gamma f(v) \int_0^\infty N_3(v') dv' \end{aligned} \quad (8)$$

The modified rate equations can be reduced to integral equations for the level populations, and these can be solved numerically in conjunction with the gain equation (4) to find the gain coefficient $g(v)$ as a function of laser spectral intensity $I(v)$ and collisional frequency Γ . The effects can be illustrated fairly easily; Fig. 1 shows normalized gain coefficient profiles assuming for illustrative purposes a Voigt intrinsic line profile with a 35 mÅ inhomogeneous Gaussian component and a 5 mÅ homogeneous Lorentzian component, for several values of the laser intensity and the collisional redistribution rate. The laser is assumed to have a FWHM of 5 mÅ, and is peaked at line center (206.38 Å). $\Gamma = 0$ in Fig. 1(a), $\Gamma = \Delta\nu_H$ in Fig.

1(b) and $\Gamma = 5 \Delta\nu_H$ in Fig. 1(c), where $\Delta\nu_H$ is the homogeneous Lorentzian width in frequency units. The spectral hole-burning evident in Fig. 1(a) is clearly diminished in Fig. 1(b) and becomes almost invisible over the laser intensity range investigated in Fig. 1(c). As Γ increases, spectral hole burning is effectively

Figure 1. Normalized gain vs. wavelength for various ratios of peak laser intensity/saturation intensity ($\alpha = I(v_0)/I_{sat}$); the velocity changing collision rate is equal to zero in (a), equal to the homogeneous line width in (b), and equal to five times the homogeneous line width in (c).

delayed until farther into saturation; this can be understood to result because the inhomogeneous nature of the Doppler broadening component does not become evident until the stimulated emission/absorption rate induced by the laser field becomes larger than the velocity-changing collision rate (the stimulated rates will essentially set the time-scales with which to compare Γ once they become larger than other transition rates, i.e. once the laser saturates). Since spectral hole burning is the cause of inhomogeneous saturation rebroadening in A.S.E. lasers (whether or not a dip actually occurs in the gain profile), it is clear that a collisional redistribution rate approximately equal to or larger than the homogeneous lifetime width will delay inhomogeneous saturation rebroadening, thus partly homogenizing the Doppler component of the intrinsic line profile.

Inclusion of collisional redistribution effects in A.S.E. laser line-transfer models is difficult because the rate equations for the level populations then become complex and must be solved numerically. The problem is further complicated with the addition of a laser line transfer equation, required to calculate the laser spectral intensity and line profile, and by the necessarily self-consistent solutions to the population rate equations and the line transfer equation. However, the inclusion of collisional redistribution effects in the usual models¹³ is in principal straightforward, and presents only numerical difficulties.

We note that our treatment of collisional redistribution is entirely equivalent to that presented in ref. 15. That work is based on an un-closed two-level rate equation model, but the collisional redistribution modifications in eqs. (47) and (48) of that work are equivalent to eq. (8) above when e.g. $N_3(v)$ is written as $N_3 f_3(v)$, so that the integral in eq. (8) is written as N_3 . Finally, we note that collisional

redistribution effects may be relevant to other A.S.E. laser line width measurements, e.g. those of ref. 23, where experimental measurements showed only slight inhomogeneous saturation re-broadening, in apparent disagreement with calculations¹³. This was also noted in ref. 15, and that author later re-examined²⁴ the experimental data of ref. 23, finding excellent agreement when collisional re-distribution effects are taken into account.

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