Mechanisms for creating population inversions in Ne-like ions

William H. Goldstein, Barbara L. Whitten, Andrew U. Hazi, and Mau H. Chen
High-Temperature Physics Division, Lawrence Livermore National Laboratory, University of California, Livermore, California 94550

(Received 15 April 1987)

We investigate the effects of inner-shell ionization of sodiumlike ions on the kinetics of neonlike selenium, using a simple model that separates the calculation of the excited-state kinetics from that of the ionization balance. Our model includes collisional and radiative transitions between the neonlike states, inner-shell ionization of sodiumlike states, and dielectronic recombination from fluorinelike ions. By examining the relative importance of these various population mechanisms, we demonstrate that inner-shell ionization of sodiumlike ions has a significant effect on the inversion kinetics of neonlike selenium, although it is not sufficient alone to account for the discrepancy with experimental observation.

INTRODUCTION

Neonlike ions have long been recognized as candidates for soft-x-ray laser schemes because of their energy-level structure. The $2s^22p^53s$ excited states can decay to the ground state by fast dipole-allowed radiative transitions, while the higher-energy $2s^22p^53p$ states can radiate to the ground state only by much slower electric quadrupole transitions. So the possibility of creating a population inversion between the $2s^22p^53p$ and $2s^22p^53s$ excited states was recognized very early.[1]

In recent years, experiments in laser-produced plasmas have demonstrated amplified spontaneous emission in neonlike selenium,[2] yttrium,[2] molybdenum,[3] and copper.[4] These experiments have also demonstrated that our understanding of the kinetics of neonlike ions is incomplete. In the first successful experiment,[2] time-integrated gains of 5.5 ± 1 cm^{-1} were measured for two $J=2$-$J=1$ transitions at 206.3 and 209.6 Å in selenium, in reasonable agreement with the predicted gains[5] of 4 cm^{-1}. However, the $J=0$-$J=1$ transition at 182.4 Å, which was predicted[5] to show a gain of 10 cm^{-1}, did not appear to be amplified significantly. The yttrium results from the same experiment are ambiguous, due to a line coincidence between the $J=0$-$J=1$ and one of the $J=2$-$J=1$ transitions. A later experiment in molybdenum,[3] however, showed results similar to selenium; gains of 4 cm^{-1} were measured for the two $J=2$-$J=1$ transitions, while the $J=0$-$J=1$ transition analogous to that at 182.4 Å in selenium was not significantly amplified. (The other $J=0$-$J=1$ transition at 106.4 Å, however, displayed a measured gain of 2.2 cm^{-1}, in reasonable agreement with the prediction of 3.5 cm^{-1}.) The very recent results in copper[4] do not display this same anomaly; here comparable gains of ≈ 2 cm^{-1} have been measured for all three lines, analogous to the 182.4-, 206.3-, and 209.6-Å transitions in selenium.

Initial efforts to model the neonlike soft-x-ray laser[5-7] assumed that the most important population mechanism was electron collisional excitation from the neonlike ground state, and most models considered only that process. Subsequent attempts to explain this discrepancy[8-10] have concentrated on including recombination from fluorinelike ions in the model of the neonlike population kinetics. In particular, dielectronic recombination has been shown to have a significant effect,[9,10] although not enough to fully explain the experimentally observed gains, if reasonable assumptions are made about the plasma conditions. It has also been suggested[11] that plasma effects on the line profiles of the amplified transitions might provide an explanation.

In this paper we investigate another method of creating the population inversion–inner-shell ionization of sodiumlike ions. Calculations by Sampson and Zhang[12] in the accompanying paper show that the rates of inner-shell ionization by electron impact are large enough to be competitive with the rates of electron-impact excitation from the neonlike ground state. We examine the efficiency of inner-shell ionization as a mechanism for creating a $3p$-$3s$ population inversion in neonlike selenium by constructing a simple model that separates the calculation of the excited-state kinetics from that of the ionization balance. This model provides an assessment of the relative importance of inner-shell ionization, collisional excitation, and dielectronic recombination as a function of ionization balance.

We demonstrate that inner-shell ionization can indeed produce a population inversion, although this process is only about half as efficient as excitation. We also show that ionization from sodiumlike ions heavily favors the $J=2$-$J=1$ inversion over the $J=0$-$J=1$. With reasonable assumptions about the relative abundance of sodiumlike, neonlike, and fluorinelike ions, however, the magnitude of the effect is not large enough to fully explain the experimentally observed gains in neonlike selenium.

THE MODEL

For neonlike selenium, we use a 36-level model which contains all the $2s^22p^53l$ and $2s2p^63l$ excited states. Collisional and radiative processes connect all these levels. Rates into the neonlike excited states from either the neonlike $2s^22p^6$ ground state, the sodiumlike $2s^22p^63l$ states, or the fluorinelike $2s^n2p^m$ $(n+m=7)$ states, are treated as source terms. We thus have

$$Rn = k_E N_{\rm Ne} + \sum_i k_I^i N_{\rm Na}^i + \sum_j k_R^j N_{\rm F}^j \ , \quad (1)$$

where n is the population vector of neonlike excited states; R is the matrix of collisional and radiative rates connecting these levels; the diagonal terms contain the losses from each level, including collisional deexcitation and radiative decay. The terms on the right-hand side of Eq. (1) form a column vector of fixed sources. The indexes i and j run over the sodiumlike $2s^22p^63l$ and the fluorinelike $2s^n2p^m$ states, respectively. k_E is the vector of rate coefficients for collisional excitation from the neonlike ground state,

$$2s^22p^6+e \Longrightarrow \begin{cases} 2s^22p^53l+e \\ 2s2p^63l+e \ , \end{cases} \quad (2)$$

the k_I^i are vectors of rate coefficient for inner shell ionization from sodiumlike ions,

$$2s^22p^63l+e \Longrightarrow \begin{cases} 2s^22p^53l+2e \\ 2s2p^63l+2e \ , \end{cases} \quad (3)$$

and the k_R^j are vectors of rate coefficients for dielectronic recombination from fluorinelike ions,

$$\left.\begin{matrix} 2s^22p^5+e \\ 2s2p^6+e \end{matrix}\right\} \Longleftrightarrow \left\{\begin{matrix} 2s^22p^43l3l' \\ 2s2p^53l3l' \\ 2p^63l3l' \end{matrix}\right\} \Longrightarrow \begin{cases} 2s^22p^53l+h\nu \\ 2s2p^63l+h\nu \ . \end{cases} \quad (4)$$

We have included only dielectronic recombination in this model, neglecting radiative and three-body recombination. This is a reasonable approximation because at the plasma conditions we are interested in, dielectronic recombination is the dominant recombination mechanism. (In the following we will suppress the indices i and j, and the ion densities, unless otherwise noted, will refer to total ion densities; i.e., $N_{\rm Na} = \sum_i N_{\rm Na}^i$, $N_{\rm F} = \sum_j N_{\rm F}^j$.)

Since Eq. (1) is linear in the ion densities $N_{\rm Ne}$, $N_{\rm Na}$, and $N_{\rm F}$, we can solve this steady-state set of rate equations separately for each source term, and calculate the relative inversion density for the transitions $U \Longrightarrow L$ according to

$$y_{UL}^k = (N_k)^{-1}(n_U^k - g_U n_L^k / g_L) \ , \quad (5)$$

where g_U and g_L are the statistical weights of the upper and lower states, respectively. The gain coefficient per source ion, for the kth species is

$$a^k = \alpha^k / N^k = \sigma_{UL} y_{UL}^k \ , \quad (6)$$

where σ_{UL} is the stimulated emission cross section for the transition $U \Longrightarrow L$. For a particular ionization balance, the total gain coefficient for the transition is then

$$\alpha = \sum_k N^k a^k = N_i \sum_k a^k f_k \ , \quad (7)$$

where N_i is the total ion density and f_k is the fraction of the kth ion species.

The approximation just described greatly simplifies the kinetic modeling and allows us to separate the calculation of the excited-state kinetics from that of the ionization balance. Since the ionization balance of the x-ray laser plasma is not well understood theoretically and has not been completely characterized experimentally, it is useful to be able to investigate the excited-state kinetics independently. This approximation of course neglects the effect of neonlike excited-state processes on the distribution of population among the multiple ground states of the sodiumlike and fluorinelike ions. But, since we are concerned only with the $3p$-$3s$ inversion density, and since the fraction of ions in any neonlike excited state is small, this feedback is of negligible importance, for reasonable plasma conditions.

The collisional and radiative rates connecting the 37 lowest states of neonlike selenium were calculated using the collisional-radiative code package of Klapsich and Bar-Shalom.[13] The dielectronic recombination rates of fluorinelike ions were calculated in the isolated resonance approximation, using the multiconfigurational Dirac-Fock model with intermediate coupling.[14]

We have used two different sets of inner-shell ionization cross sections. One set was calculated by Sampson and Zhang[12] using a Z-scaled hydrogenic model and the Coulomb-Born exchange approximation. The other set was obtained by extrapolating the results of Younger[15] for Mg^+, Al^{2+}, P^{4+}, and Ar^{7+}, calculated using the distorted-wave exchange approximation. As seen in Fig. 1, the two cross sections are in good agreement except at very high energy, where they are normalized to somewhat different photoionization cross sections. For the

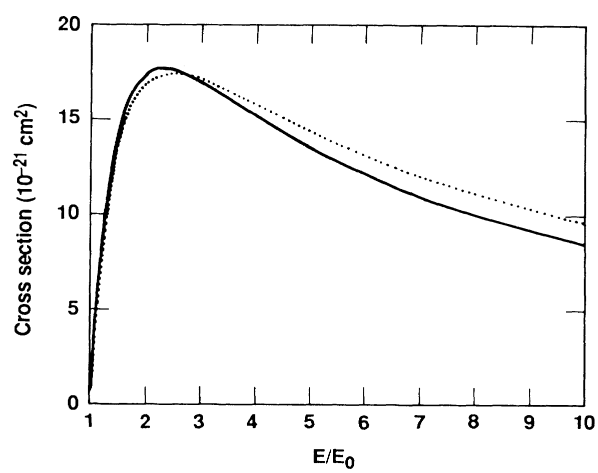

FIG. 1. Total ionization cross section (cm^{-2}) vs energy (threshold units) for the process $2s^22p^63s+e \Longrightarrow 2s^22p^53s+2e$ in selenium. Solid line: scaled hydrogenic cross section (Ref. 12). Dotted line: distorted wave cross section extrapolated from those in Ref. 15.

temperatures we are interested in, the two sets of rate coefficients are in excellent agreement.

To obtain the ionization rate into a specific neonlike excited state, the total rate for ionizing a 2s or 2p electron must be multiplied by the appropriate branching ratio. Recently, Sampson[16] has derived general formulas for these branching ratios, which in the case of sodiumlike ions give the simple result

$$k_I^{\rm det}=(2J+1)/(2j+1)k_I \ , \tag{8}$$

where $k_I^{\rm det}$ is the ionization rate coefficient into a particular neonlike excited state, k_I is the total ionization rate coefficient for a 2s or 2p electron, J is the total angular momentum of the final neonlike excited state, and j is the angular momentum of the $3l$ spectator electron. This approximation assumes that the spectator electron is unaffected by the ionization of the inner-shell electron, and ignores two-electron transitions, such as

$$2s^22p^63s_{1/2}+e \Longrightarrow 2s^22p^53p_{3/2}+2e \ , \tag{9}$$

which we expect to have negligible rates. Transitions in which the total angular momentum, but not the orbital angular momentum, of the spectator electron changes, such as

$$2s^22p^63p_{1/2}+e \Longrightarrow 2s^22p^53p_{3/2}+2e \ , \tag{10}$$

are correctly accounted for in our model, provided that the population within the $3l$ subshell is statistically distributed. To test this assumption, we calculated the populations of the sodiumlike states using the 12-level model described below. The results are given in Table I. Clearly, this assumption is excellent for densities above $n_e=3\times10^{20}$ cm^{-3}.

Using the neonlike model and the rates just described, we solved Eq. (1) separately for each source term, and then evaluated the gain per source ion from Eqs. (5) and (6). In order to use Eq. (1), it is necessary to know the relative populations of the five sodiumlike $2s^22p^63l$ states, and the three fluorinelike $2s2p^6$ and $2s^22p^5$ states. These were obtained from two independent calculations using the simplified models described below.

The relative populations of the sodiumlike states were computed with a collisional-radiative model including the 12 levels obtained from the configurations $2s^22p^63l$ and $2s^22p^64l$. All radiative and collisional rates were computed using the Klapisch–Bar-Shalom codes.[13] Ionization and recombination were not included. The only such process that might be important at relevant plasma conditions is dielectronic recombination from the neonlike ground state. But the relative dielectronic recombination rates into the different $2s^22p^63l$ states are very nearly statistical and will not perturb the already nearly statistical distribution obtained from collisional excitation alone.

The relative populations of the fluorinelike states were determined with a collisional-radiative model containing the lowest 113 fine-structure levels arising from configurations of the form $2s^p2p^q3l^r$, with $r=0,1$ and $p+q+r=7$. Again, the Klapisch–Bar-Shalom codes[13] were used to compute collisional and radiative rates, and ionization and recombination processes were neglected.

TABLE I. Ratio of populations within 3p and 3d subshells for sodiumlike selenium versus electron density, using the collisional-radiative model described in the text. Results shown are for $T_e=1000$ eV, although the temperature dependence is weak.

n_e (cm^{-3})	$[3p_{1/2}]/[3p_{3/2}]$	$[3d_{3/2}]/[3d_{5/2}]$
1×10^{18}	0.82	1.00
1×10^{19}	0.68	0.68
3×10^{19}	0.59	0.65
1×10^{20}	0.54	0.65
3×10^{20}	0.51	0.66
1×10^{21}	0.50	0.66
1×10^{22}	0.50	0.67

RESULTS

First, we studied the relative importance of collisional excitation of neonlike ions and inner-shell ionization of sodiumlike ions as population mechanisms for the excited states of neonlike selenium. The total gain coefficients of the relevant $2p^53p$-$2p^53s$ transitions were evaluated from Eq. (7) by assuming $N_{\rm F}=0$, treating the ratio of sodiumlike to neonlike ions as a parameter, and taking the relative populations of sodiumlike states from the 12-level model described above. The results are shown in Fig. 2, where the gain coefficients of several transitions are plotted as a function of the ratio of sodiumlike to neonlike ions. Plasma conditions are taken to be $T_e=1000$ eV, $n_e=5\times10^{20}$ cm^{-3}, and $T_i=400$ eV. The total density of neonlike plus sodiumlike ions is assumed to be 6×10^{18} cm^{-3}.

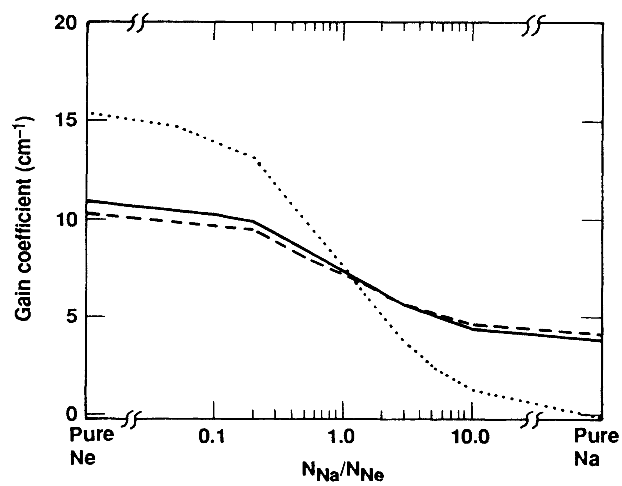

FIG. 2. Gain coefficient (cm^{-1}) of $2p^53p$-$2p^53s$ transitions in neonlike selenium vs ratio of sodiumlike to neonlike ions. The plasma conditions are taken to be $T_e=1000$ eV, $n_e=5\times10^{20}$ cm^{-3}, $T_i=400$ eV, $N_{\rm Ne}+N_{\rm Na}=6\times10^{18}$ cm^{-3}. Dotted line: $J=0$-$J=1$ transition at 182.4 Å. Solid line: $J=2$-$J=1$ transition at 209.6 Å. Dashed line: $J=2$-$J=1$ transition at 206.3 Å.

If one compares the gains at the far left of the figure (collisional excitation only) with those at the far right (inner-shell ionization only), it is clear that inner-shell ionization is about one-half to one-third as effective at creating an inversion as is collisional excitation. This behavior can be traced to the fact that, although ionization into any particular level tends to proceed faster than collisional excitation,[12] the difference in feeding rates for the upper and lower laser levels is more favorable to the population inversion in the latter process. Of course, for both pumping mechanisms, it is the selective depopulation of the lower level through the dipole transition to the ground state that sustains the inversion. It should also be pointed out that the present calculation does not include the effects of optical depth in the neonlike 3-2 lines. Including this effect may reduce the calculated gain coefficients at high N_{Ne}.

It is also clear from Fig. 2 that the substantial amplification of the $J=2$-$J=1$ transitions persists in a plasma containing only sodiumlike ions, while the $J=0$-$J=1$ amplification does not. This is not unexpected, since inner-shell ionization favors states with large statistical weights.

We next wish to investigate the relative importance of the three population mechanisms as a function of temperature. Figure 3 shows a^k, the gain per source ion density for each of the three ion species, for the $J=2$-$J=1$ transition at 209.6 Å, and for the $J=0$-$J=1$ transition at 182.4 Å. It is clear that for the $J=0$-$J=1$ transition collisional excitation is the only important mechanism for producing inversion. The $J=2$-$J=1$ transition shows significant gain from either recombination or ionization, in addition to collisional excitation.

In order to fully assess the relative importance of the three mechanisms, we must multiply a^k by N_k, the appropriate source ion density. We must therefore determine the ionization balance of the selenium x-ray laser plasma. This is an extremely difficult problem because the plasma is not in steady state, and there are temporal and spatial gradients present. There is no direct measurement of the ionization balance, and the predictions of different time-dependent codes do not agree.

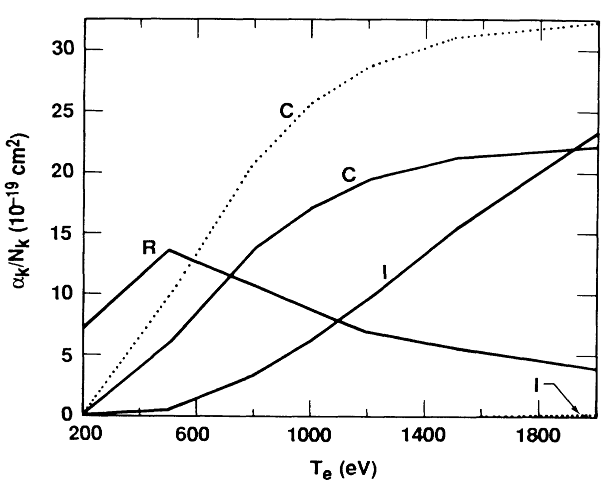

FIG. 3. Gain coefficient per source ion density (cm^2) of $2p^53p$-$2p^53s$ transitions in neonlike selenium vs electron temperature (eV) for $n_e=5\times10^{20}$ cm^{-3}. Dotted lines: $J=0$-$J=1$ transition at 182.4 Å. Solid lines: $J=2$-$J=1$ transition at 209.6 Å. I: gain per sodiumlike ion (from inner-shell ionization). C: gain per neonlike ion (from collisional excitation). R: gain per fluorinelike ion (from dielectronic recombination). (Curve R for the $J=0$-$J=1$ transition is too small to be seen.)

Steady-state calculations of the ionization balance indicate that the neonlike ion fraction reaches its peak between 600 eV (Ref. 17) and 1000 eV (Ref. 18) for $n_e=5\times10^{20}$ cm^{-3}. Thus the neonlike ion density and the collisional excitation rate are both large for roughly the same plasma conditions. In contrast, the fluorinelike fraction peaks at higher temperatures, where the recombination rate is small; at lower temperatures the sodiumlike fraction will increase, but the collisional ionization rate falls. Therefore, if the plasma is in steady state, collisional excitation will be a more effective mechanism for creating a population inversion in neonlike ions than either dielectronic recombination or inner-shell ionization.

Because conditions in the x-ray laser plasma are changing rapidly relative to the overall ionization and recombination rates, the ionization balance is probably not in steady state.[19] In such a case, the above argument is not valid, and it is possible to construct scenarios[8] in which, for example, a mostly fluorinelike plasma cools rapidly and creates a population inversion due to recombination.

CONCLUSIONS

We have shown that inner-shell ionization from sodiumlike ions has a significant effect on the excited-state kinetics of neonlike ions, and is an effective mechanism for creating a $2p^53p$-$2p^53s$ inversion. How effective it is, of course, depends on the relative abundance of sodiumlike ions in the plasma. At present we cannot calculate the ionization balance with any confidence. However, we see from Fig. 2 that unless the ratio of sodiumlike to neonlike ions is larger than 1, ionization will not reduce the expected gain of the $J=0$-$J=1$ transition enough to explain the experimental results.

So far all efforts to model the excited-state kinetics of neonlike selenium have failed to find a process which, by itself, can reduce the gain of the $J=0$-$J=1$ transition at 182.4 Å enough to bring it into line with the experimental results. It is possible, however, that a number of processes, all favoring the population of the $J=2$ upper levels, combine to increase the gain on the $J=2$-$J=1$ transitions relative to the $J=0$-$J=1$. In this context, the effect of inner-shell ionization of sodiumlike ions is a significant population mechanism.

Inner-shell ionization may be more significant in explaining the same anomaly in molybdenum.[3] Simple estimates of the ionization balance from time-integrated x-ray spectra show that the molybdenum plasma is less highly ionized than selenium, and the ratio of sodiumlike to neonlike ions may be as high as 0.5.[20] In addition, the ratio of ionization to excitation rates is more favorable at higher Z.[12]

The present results also show that, because the $2p^53p$ $J=2$ levels are populated by dielectronic recombination and inner-shell ionization as well as collisional excitation, inversion of the $J=2$-$J=1$ levels will occur even if the plasma is over- or underionized, so that neonlike ions are not the most abundant species. This result helps to explain the experimental observation[21] that the strongly amplified $J=2$-$J=1$ lines at 206.3 and 209.6 Å in selenium occur over a wide range of plasma conditions.

ACKNOWLEDGMENTS

The authors would like to thank Rosemary Walling for profitable discussions of kinetic and modeling issues. We are also grateful to Professor Douglas H. Sampson and Dr. Honglin Zhang for useful discussions, and for providing us with their results prior to publication. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

[1]A. N. Zherikhin, K. N. Koshelev, and V. S. Letokhov, Kvantovaya Elektron. (Moscow) **3**, 152 (1976) [Sov. J. Quant. Electron **6**, 82 (1976)].

[2]D. L. Matthews *et al.*, Phys. Rev. Lett. **54**, 110 (1985).

[3]B. J. MacGowan *et al.*, J. Appl. Phys. **61**, 5243 (1987).

[4]T. N. Lee, E. A. McLean, and R. C. Elton, Phys. Rev. Lett. **59**, 1185 (1987).

[5]M. D. Rosen *et al.*, Phys. Rev. Lett. **54**, 106 (1985).

[6]A. V. Vinogradov and V. N. Shlyaptsev, Kvantovaya Elektron. (Moscow) **7**, 1319 (1980) [Sov. J. Quantum. Electron. **10**, 754 (1980)], and references therein.

[7]U. Feldman, A. K. Bhatia, and S. Suckewer, J. Appl. Phys. **54**, 2188, (1983); U. Feldman, J. F. Seely, and A. K. Bhatia, *ibid.* **56**, 2475 (1984).

[8]J. P. Apruseze, J. Davis, M. Blaha, P. C. Kepple, and V. L. Jacobs, Phys. Rev. Lett. **55**, 1877 (1985).

[9]B. L. Whitten, A. U. Hazi, M. H. Chen, and P. L. Hagelstein, Phys. Rev. **33**, 2171 (1986).

[10]P. L. Hagelstein, M. D. Rosen, and V. L. Jacobs, Phys. Rev. A **34**, 1931 (1986).

[11]H. Griem, Phys. Rev. A **33**, 3580 (1986).

[12]D. H. Sampson and H. Zhang, preceding paper, Phys. Rev. A **36**, 3590 (1987).

[13]M. Klapisch and A. Bar-Shalom (private communication).

[14]M. H. Chen, Phys. Rev. A **34**, 1079 (1986).

[15]S. M. Younger, Phys. Rev. A **24**, 1272 (1981).

[16]D. H. Sampson, Phys. Rev. A **34**, 986 (1986).

[17]M. D. Rosen (private communication).

[18]Y. T. Lee (unpublished).

[19]See, for example, W. H. Goldstein, R. S. Walling, J. Bailey, M. H. Chen, R. Fortner, M. Klapisch, T. Phillips, and R. E. Stewart, Phys. Rev. Lett. **58**, 2300 (1987).

[20]T. A. Phillips (private communcation).

[21]D. L. Matthews (private communication).