Self-Organization, Broken Symmetry, and Lasing in an Atomic Vapor: The Interdependence of Gratings and Gain

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We describe a novel optical gain mechanism observed in a strongly driven Doppler-broadened atomic vapor. Gain and self-stabilizing laser oscillations are observed in a direction which is *counterpropagating with respect to the traveling wave pump*. Using Bragg diffraction, we show that the underlying gain-lasing mechanism is directly tied to a spontaneously formed optical wavelength grating. We suggest that this grating arises from a self-organization or self-bunching of the atoms, an effect which is mediated by recoil just as in the free electron laser. [S0031-9007(96)00949-0]

The generation of coherent short-wavelength radiation from relativistic particle beams often requires the efficient direction reversal of optical radiation such as occurs in the free-electron laser. One efficient strategy for implementing this idea is to backdiffract pump radiation from a moving atomic grating. Recently, it has been shown that the efficiency of such a grating can be greatly enhanced by using a coupling of the internal and the external states of the atoms—that is, by taking advantage of atomic recoil. This enhancement process has been described in two complementary pictures. In one picture [1], the optical forces generated by the interference of a strong pump and counterpropagating fluctuation ("probe") cause the spontaneous, collective organization of the atoms into a density grating. The pump field backdiffracts from this grating into the probe resulting in amplification of the probe. In turn, the increase in the probe field strength causes an increase in the forces and hence in the bunching, a cycle which provides strong cooperative feedback in the grating formation process. In this picture, the probe represents a self-consistent field which together with the grating grows in strength, giving rise to an instability in the vapor. A laser based on this gain mechanism has been proposed and is referred to as the collective atomic recoil laser (CARL) [1]. In a complementary [2,3] picture, the gain is attributed to a single particle process associated with stimulated transitions between different atomic velocity groups, transitions which are referred to as recoil induced resonances and are related to stimulated optical Compton scattering. Indeed, in a set of elegant experiments, Courtois *et al.* [3] have recently reported the first experimental observation of these resonances in a laser cooled atomic cesium vapor and they have described measurements of probe gain which result from these recoil effects. In either

picture, the similarity of this atomic gain mechanism to that of the free-electron laser is clear [1,3,4] and this correspondence has provoked significant interest [5].

In this Letter, we present direct experimental evidence that a highly efficient, short wavelength grating can spontaneously form in a strongly pumped atomic sodium vapor *without the introduction of a counterpropagating probe*. We show that with increasing density, this grating can have a dramatic effect on reverse gain, so dramatic that in the presence of a surrounding cavity, self-starting continuous reverse lasing oscillations occur [6]. In previous experiments [3,4,7,8] an external probe, counterpropagating with respect to the pump, was introduced and any grating formation was therefore induced by the interfering pump and probe fields. In particular [4], the backreaction of the amplified probe on grating formation was weak and a collective instability was not indicated. By contrast, in our experiments, the spontaneous formation of the grating and the demonstrated link of this grating to the lasing mechanisms are clear signatures of a collective instability in the atom + pump field system—signatures which characterize the CARL process. Furthermore, our observations of the spontaneous grating are particularly remarkable given the large thermal velocity spread of the lasing medium [4].

The essential experimental apparatus consisted of a thermal sodium source and two frequency stabilized ring dye lasers. Four different source configurations were used: a vapor cell with either a 1 or 3 cm path length (measured Doppler width $\Delta_{\text{Dop}} \sim 1.5$ GHz FWHM), a high flux multicollimator beam with a 5 mm diameter/interaction length (measured $\Delta_{\text{Dop}} \sim 1$ GHz, centered about $\Delta = 0$) and a high-flux nozzle beam (1 mm diameter). The beams were not collimated and the pump field was nearly perpendicular to the beam axis. Each source has been used to implement

a reverse laser and each has exhibited similar properties. The pump laser was tuned near to the $3S_{1/2}(F = 2) \rightarrow 3P_{3/2}(F' = 1, 2, 3)$ transition (the D_2 line) and was circularly polarized ($\equiv \sigma+$). When stabilized, the laser linewidth was ~1 MHz although similar results were obtained when the lasers were left nonstabilized (~20 MHz linewidth). All frequencies were accurately determined by reference either to a Doppler-free saturated absorption cell or to a highly collimated atomic beam.

The geometry of the system where stable lasing was observed is shown in Fig. 1(a): a folded ring cavity containing the sodium source. The vapor is excited nearly on axis by the strong traveling wave pump. Both pump and cavity were mode matched and focused to ~300 μm (FWHM) at the interaction region using two symmetrically placed 50 cm focal length lenses (the Rayleigh length in this case is ~12 cm $\gg$ source dimensions). The pump angle was <10 mrad from the cavity axis. Pump beam intensities from 100 to 1000 W/cm^2 were used corresponding to on resonance Rabi frequencies $\Omega_R \lesssim 2.5$ GHz.

The first new observation is that above a critical atom density (N_T) and pump Rabi frequency (Ω_R) we observed self-starting, self-sustaining laser oscillations in a direction opposite to the direction of propagation of the pump. It is this reverse oscillation which is the subject of this Letter. Forward oscillations could also be observed using this system both in the presence and absence of the reverse oscillations. Most important, however, is that reverse oscillations could be observed *in the absence of any forward oscillations*. When present, the forward oscillation had all of the properties of the three-photon (so-called Raman) gain component of the Mollow laser [9–11] (a Raman laser [12] could also be observed). The reverse oscillations, however, are not explained by any previously described Mollow or Raman mechanisms. Inspired by the CARL model, we used Bragg scattering [see Fig. 1(b)] to identify a short wavelength grating in the atomic vapor and to show that the presence of this grating is intimately tied to the observed gain. This direct evidence of a grating represents our second new observation.

Consider first the reverse oscillations. In Fig. 2 we show a typical frequency spectrum of this radiation for $\Delta_{\text{pump}} \approx -500$ MHz. The output is redshifted from the pump by ~2.5 GHz, is ~1.8 GHz wide, and on close inspection it is observed to be composed of a series of discrete modes at the cavity spacing. This spectrum is not characteristic of the Raman laser [12]: it is broad and shifted by 2.5 GHz (not the 1.77 GHz splitting of the Na ground state). This oscillation is also not trivially ascribed to the Mollow gain [9] because (1) Doppler averaging is known to substantially suppress the reverse Raman gain feature and (2) the observed frequency shift excludes the Rayleigh gain, a mechanism which does survive the Doppler averaging [11]. However, the spectrum is consistent with predictions from the CARL theory, a model which, for our parameters, predicts a frequency shift ~2–2.5 GHz as given by the collective recoil bandwidth $\omega_r \rho$ [1]. The sign of the shift indicates that
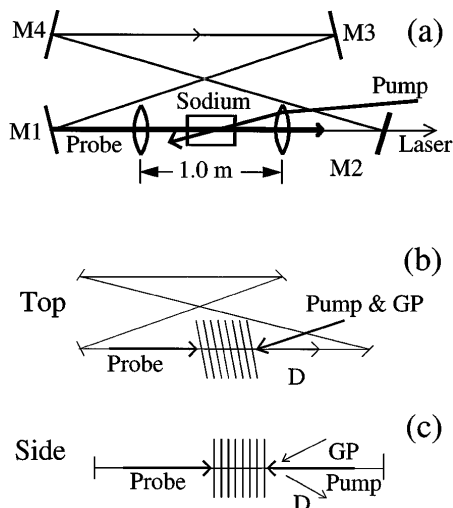

FIG. 1. (a) Ring cavity. Mirrors M1 and M2 had reflection coefficients $R = 0.98$ and served as output couplers while the M3 and M4 had $R > 0.999$. All source windows and lenses were antireflection coated ($R < 0.005$). (b) Top view of ring showing geometry of grating probe (GP) and pump (P) beams. (c) Side view of ring showing the diffracted beam (D).
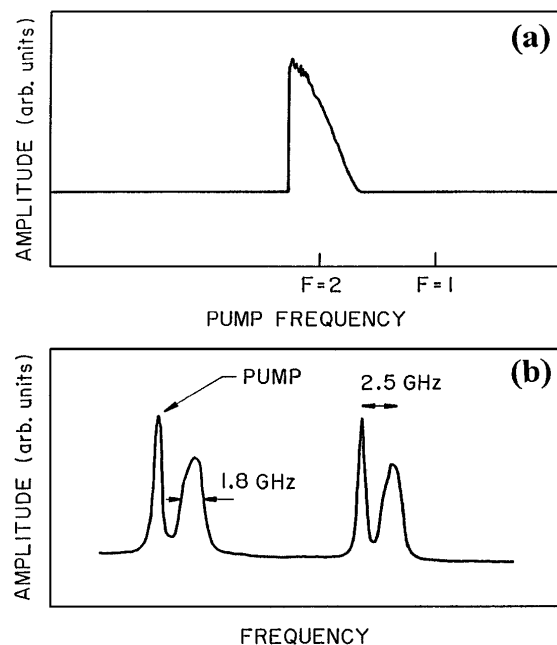

FIG. 2. (a) Reverse oscillation intensity as a function of pump detuning normalized to the pump intensity for the 1 cm cell (at 300 °C). The full width at half maximum is 0.7 GHz. Frequency increases to the left. (b) The output spectrum of the reverse oscillation for $\Omega_R \sim 1.5$ GHz and pump detuning $\Delta_{\text{pump}} \approx -500$ MHz measured using a 15 GHz free spectral range Fabry Pérot interferometer. Frequency increases to the right.

the grating is copropagating with the pump in qualitative agreement with [3] except that here the magnitude of the shift is not Δ_{Dop} but instead $\omega_r \rho$.

A typical plot of reverse laser output intensity as a function of Δ_{pump} is also shown in Fig. 2. The peak occurs at ~500 MHz to the red of the $F = 2 \rightarrow F' = 0, 1, 2, 3$ transition. The laser output was of the same polarization as the pump and the intensity was observed to decrease rapidly for pump detuning $\Delta_{\mathrm{pump}} \lesssim -500$ MHz due to the onset of forward Mollow lasing which competes for pump energy. For blue detunings the lasing rapidly ceases due to pump absorption and pump self-focusing effects. When the $\sigma+$ pump was tuned resonant with the D_1 transition ($3S_{1/2} \rightarrow 3P_{1/2}$), lasing was not observed. We attribute this to the lack of a closed two-level system on this transition and to competition with the strong forward D_1 Raman laser [12]. The peak reverse intracavity fluence (normalized to pump intensity) was 0.03, 0.08, and 0.30 for the 1 cm cell, the 1 mm beam, and the 3 cm cell, respectively. For each setup, a clear pump intensity threshold was observed which was $\gtrsim 250$ W/cm^2. The existence of a pump threshold and the observed values are in good agreement with the CARL theory. Moreover, for both cells and for the 1 mm beam, the density threshold n_T for lasing occurred for source temperatures of within ten degrees of 340 °C with $n_T \sim 130$ atoms/$\lambda_{\mathrm{optical}}^3$. For the multicollimated beam, a similar source threshold temperature was observed; however, low density absorption measurements scaled to 340 °C indicated $n_T \sim 50$ atoms/$\lambda_{\mathrm{optical}}^3$. To reconcile this, we note that for the open geometry of the cells [13] and the 1 mm beam, the n_T is not as easily determined as for the multicollimated beam. The important point is that in each case n_T is in good agreement with the threshold $n_T = 40$ atoms/$\lambda_{\mathrm{optical}}^3$ calculated by equating the CARL bandwidth $\omega_r \rho$ [1,14] with the inhomogeneous linewidth. In the CARL theory, the gain is dominated by the collective bandwidth. Here, since $\omega_r \rho > \Delta_{\mathrm{Dop}}$, Doppler averaging should not destroy the grating enhanced gain [15]. Furthermore, the fact that the lasing threshold depends more on *density* than on the total number of atoms (i.e., the interaction path length) is in agreement with CARL theory.

To directly observe the grating, an additional grating probe (GP) laser beam was focused into the interaction region, as shown in Fig. 1(b). GP was from 4% to 25% as intense as the pump, incident at ~10 mrad out of the pump or probe plane and nearly copropagating with the pump. The GP could be derived from the pump, the reverse laser, or from a second stabilized dye laser. In this geometry we were able to observe a Bragg-matched backscattered portion of GP exiting the interaction region as shown. The Bragg scattered spot had the same polarization and frequency spectrum as the GP and scattering could be observed when the GP was both $\sigma+$ and $\sigma-$ polarized. It is important to recognize that

there can be other wave mixings in this geometry that can give rise to a grating; however, only a short wavelength grating oriented perpendicular to the lasing axis can be responsible for the observed Bragg scattered spot.

To link the grating with the reverse gain we have carried out experiments in which the external cavity was removed and lasing thereby interrupted. We used the setup shown in Fig. 1(a), except that M1 and M2 were removed. Without the cavity, coherent backscattering (lasing) was not clearly observed, so, to induce grating formation, a reverse probe was introduced which exactly overlapped the reverse lasing path [see Fig. 1(a)]. We were thereby able to measure the single-pass reverse probe gain ($I_{\mathrm{transmitted}}/I_{\mathrm{input}}$). For probe detunings for which gain was observed, a grating was simultaneously detected by Bragg scattering and we were therefore able to link this gain with the grating. Figure 3 depicts the system response as a function of reverse probe frequency. Here $\Delta_{\mathrm{pump}} \approx -1500$ MHz was chosen to maximize the observed gain. Notice that the reverse gain is reduced by ~2 when light is diffracted from the grating. Moreover, the loss in final output probe power was approximately equal to the power transferred to the diffracted beam. For Fig. 3, the GP was derived from the same laser as the pump, but was orthogonally polarized ($\sigma-$). In a different experiment using the oscillating laser, we
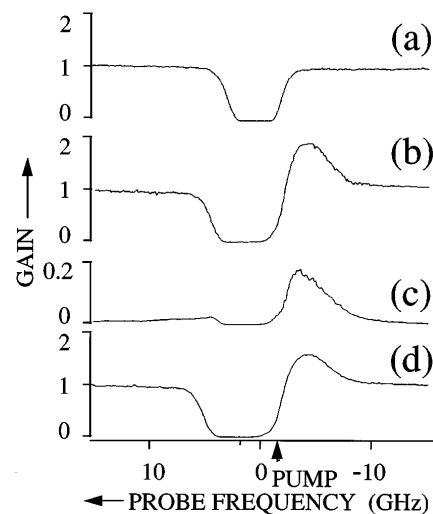
FIG. 3. The interplay of reverse gain and the grating without the ring cavity. (a) The probe absorption in the absence of a pump, (b) and (d) show the probe gain spectrum without and with the grating probe, respectively, and (c) shows the diffraction efficiency. Here $\Delta_{\mathrm{pump}} \approx -700$ MHz. In order to achieve a high signal to noise ratio, the gain was increased by increasing I_{pump}. This was accomplished by replacing the 50 cm lenses [see Fig. 1(a)] by $f = 30$ cm focal length lenses, separated by 60 cm. This reduced the pump (and probe) spot sizes to about 180 μm yielding $I_{\mathrm{pump}} \sim 800$ W/cm^2. I_{probe} was ~15 W/cm^2 and I_{GP} is 200 W/cm^2. To achieve high efficiencies, with smaller f lenses, the GP and diffracted beams were ~5 mrad from colinear.

found that for $(\sigma+)$ polarized grating probe and pump diffraction is also observed but its peak is a factor of 2 smaller than in the $\sigma-$ case. In this case attenuation of the backpropagating intracavity oscillation is also less (about 30% at peak diffraction).

To determine whether this gain reduction was due to perturbation of the grating or optical pumping, the experiment was repeated with the GP tuned to the sodium D1 transition, but with all other conditions identical. Since the D_1 transition 0.5 THz away from the D_2 transition, the GP will no longer be Bragg matched to diffract from the grating. However, the optical pumping rate out of the $F = 2$, $m_F = 2$ ground state sublevel is ~ 2 times faster for $(\sigma-)$ excitation on D_1, compared to D_2, so that optical pumping effects are enhanced. When the grating probe was $\sigma-$ polarized and tuned resonant with the same velocity group as the pump, no diffracted beam or intracavity probe attenuation was observed.

The competition between gain and grating-probe diffraction was examined as a function of atomic density. The results for the multicollimator beam are shown in Fig. 4(a). Here we show the reverse probe gain as a function of density when the GP was and was not present. Clearly as atomic density is increased, the perturbation of the grating can have an increasing effect on the reverse gain. The solid line is a guide to the eye for an $n^{1/3}$ scaling law consistent with CARL theory, normalized at unity gain. The intensity dependence of the gain was also examined and, as in the lasing case, a clear threshold was observed as shown in Fig. 4(b).

In summary, we have presented clear evidence for the spontaneous formation of an atomic grating in a strongly driven atomic vapor. We have shown that this grating is intimately coupled to a strong gain mechanism which can operate in a direction which is counterpropagating with respect to the traveling wave pump field. We have used this process to construct a CW ring laser in which grating formation and reverse oscillations arise spontaneously without any seed field. Finally, these observations are well describe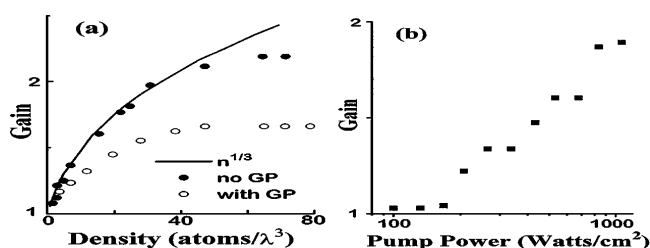d in terms of a collective instability of the atom-field system, an instability which shares many key characteristics with the collective atomic recoil laser.

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[13] The cells were heat pipes. Because no buffer gas was used there is uncertainty density uniformity. Even with 1 torr of He, no significant changes were observed.

[14] The threshold intensity scales as $c\hbar\omega_{\text{optical}}\rho n$.

[15] The radiation pressure force exerted by the pump can be neglected for short interaction times ($\tau < 1$ μs) since the recoil shift $\omega_r\Gamma\tau \ll \omega_r\rho$, the collective bandwidth.

FIG. 4. (a) Peak gain for the backpropagating probe as a function of atomic density (atoms/$\lambda^3_{\text{optical}}$) for the multicollimator source. The solid line is given by the $n^{1/3}$ CARL gain scaling scaled to fit the curve at unity gain. (b) Gain as a function of the logarithm of pump power.