Direct Observation of the Influence of Doppler-Induced Resonances on Atomic Velocities

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We report the first direct observation of the effect of Doppleron resonances on the velocity distribution of atoms in an intense optical standing wave. These observations are in reasonable agreement with the simulations based on a continued-fraction solution for the force.

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The interaction between a two-level atom and a nearly resonant light field is an interesting physical problem which has received considerable experimental and theoretical attention. An important motivation has been the understanding of the behavior of atoms in gas lasers. Theoretical work has included calculations of the interaction between a two-level atom and an intense standing wave.[1,2] These calculations predicted that the force-versus-velocity curve would display many sharp resonances referred to as Dopplerons by Kyrölä and Stenholm.[3] In this Letter, we report on the first direct observation of the effect of these Doppleron resonances[4] on an atomic velocity distribution. These observations were made using an atomic beam of sodium atoms collinear with an intense optical standing wave, where the force due to the Dopplerons is shown to exceed the maximum attainable force for purely spontaneous processes.

As is well known, the force on a two-level atom in a weak standing-wave light field is well approximated by the sum of the spontaneous forces due to the individual counterpropagating traveling waves (TW) which compose the standing wave (SW). For a weak light field with a frequency ω and a detuning Δ from the two-level transition frequency ω_0, the force-versus-velocity curve $F(v)$ for the atom has two peaks at the velocities $\mathbf{v}$ for which the Doppler shift tunes the atom into resonances with each of the two TW fields, that is, when $\pm\mathbf{k}\cdot\mathbf{v}=\Delta$. Here $\pm\mathbf{k}$ are the wave vectors of the copropagating and counterpropagating TWs, respectively. In contrast, in an intense standing wave (intensity $I \gg I_{sat}$, the saturation intensity of the two-level transition), $F(v)$ cannot be represented by a simple sum because stimulated emission processes in which the atom coherently transfers photons between the two TW fields can become very important. The force is no longer characterized merely by two peaks, but displays many narrow resonances (Fig. 2). The effects of these resonances on the evolution of the two-level density matrix were investigated theoretically by Kryölä and Stenholm.[3]

Early pump-probe experiments by Reid and Oka[5] demonstrated that the Doppleron resonances could affect the susceptibility of atoms in a gas. Their experiments measured the absorption from a weak probe field due to atoms in a strong SW pump field. The resulting spectra displayed resonances due to various interactions between the pump, the probe, and the atoms. These spectra included features associated with the velocity-tuned resonance involving three photons from the pump field. Subsequently, Minogin and Serimaa[4] analyzed the force experienced by a two-level atom in an intense SW using a continued-fraction method.[1] They pointed out that the Doppleron resonances should play an important role in determining the spatial motion of the atom.

In our experiments, we have for the first time[6] directly observed the effect of the Doppleron resonances on the atomic velocity distribution $n(v)$. We show that $n(v)$ is significantly affected by the force associated with Doppleron resonances involving as many as seven photons from the pump field (the SW). Our observations were not complicated by cross resonances between the pump and probe fields because the pump was turned off during the time that the probe was turned on. These observed $n(v)$ are found to be in good agreement with numerical simulations based on a continued-fraction expansion of the force.[3]

Several measured longitudinal velocity distributions $n(v)$ for different Δ are shown in Fig. 1. Here $n(v)$ is

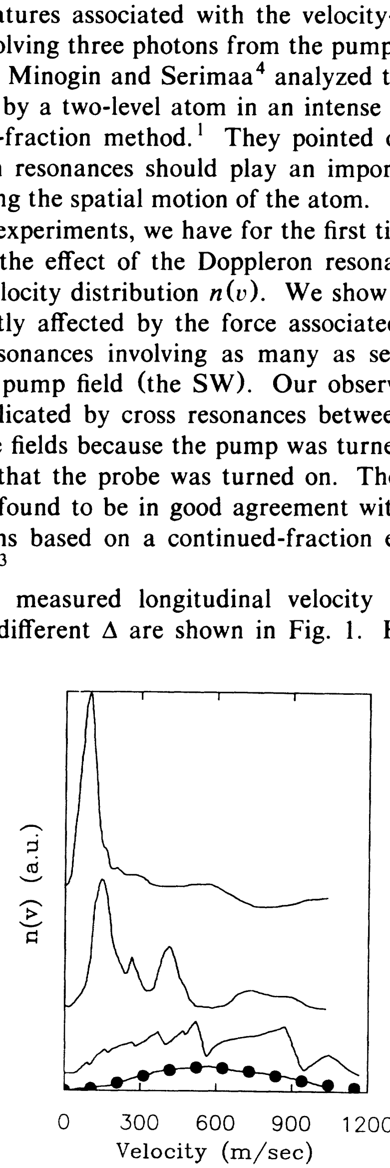

FIG. 1. Measured velocity distributions $n(v)$ for several standing-wave detunings, Δ. From the top curve down, $\Delta=-1400$, -1900, and -3200 MHz from the Na $3^2S_{1/2} \rightarrow 3^2P_{3/2}$ transition. The bottom curve is $n(v)$ in the absence of the standing wave, where the circles are the fit with a Maxwell-Boltzmann distribution. All curves are shown on the same vertical scale.

the density of atoms with velocity v parallel to $\mathbf{k}$. In order to understand these results, consider the evolution of a thermal (Maxwell-Boltzmann) distribution of atomic velocities under the influence of the acceleration curve $a(v)$ shown in the lower part of Fig. 2, where the maximum force attainable for purely spontaneous processes ($\sim 10^6$ m/sec^2) is shown by the dash-dotted line. All atoms with velocities greater than the region of the zero crossing in $a(v)$ ($v \approx 130$ m/sec) are decelerated. Atoms with velocities near the Doppleron peaks experience the largest forces and are most rapidly decelerated. Hence, the velocity groups around the Doppleron resonances are most quickly depopulated, giving rise to "holes" in $n(v)$. In the absence of collisions or diffusion, the atoms will eventually stabilize at velocities corresponding to the zeros in $a(v)$. The zeros for which $\partial a(v)/\partial v < 0$ form the set of stable equilibria for the atomic velocities and will give rise to peaks in the final $n(v)$.[7,8] If the interaction time between the atoms and the light is not long enough for all of the atoms to accumulate at the stable equilibria in velocity space, the atoms will "pile up" at velocities just below the Doppleron resonances. As $|\Delta|$ is decreased, the number of Doppleron peaks increases (cf. below) so the slowing force averaged over an optical wavelength becomes larger, and

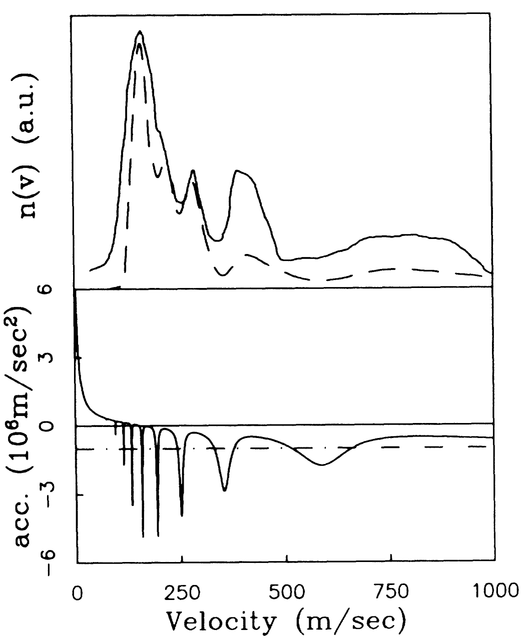

FIG. 2. Top: (solid line) measured $n(v)$ for $\Delta = -1800$ MHz and (dashed line) numerical simulation, using experimental Δ and saturation parameter $g = I/I_{sat} = 6\times 10^4$. Bottom: longitudinal acceleration $a(v)$ averaged over an optical wavelength under conditions calculated using the continued-fraction method. Calculation assumes the conditions of the top curve. The sharp peaks are the Doppleron resonances. The dash-dotted line corresponds to the acceleration due to the purely spontaneous force ($\sim 10^{16}$ m/sec^2).

hence $n(v)$ peaks at low v (upper curve in Fig. 1). In contrast, as $|\Delta|$ increases, the number of peaks decreases, the peaks move to higher velocity, the average slowing force decreases, and hence $n(v)$ resembles more closely the starting distribution with holes near the Doppleron resonances.

The experimental apparatus is very similar to that described elsewhere,[8,9] and will not be described in detail here. We only point out specific details relevant to these experiments. A standing-wave laser field was arranged collinear with a Na atomic beam. The atomic beam and the light follow the same path over the entire interaction length of 17 cm. The standing wave had a waist $w_0 \approx 0.13$ mm and a total power of 200 mW, yielding a saturation parameter $g = I/I_{sat} \approx 6\times 10^4$. Measurements were made for $-1400 < \Delta < -3400$ MHz with respect to the Na $3^2S_{1/2} \rightarrow 3^2P_{3/2}$ transition. The longitudinal velocity distribution $n(v)$ was determined by measuring the fluorescence from a weak, circularly polarized probe field which intersected the atomic beam at a shallow angle. The intersection was centered 17 cm after a 1.5-mm-diam aperture located 10 cm from the 1.5-mm aperture of the oven. The 17-cm interaction length was chosen to permit sufficient interaction time for the Doppleron resonances to burn holes in $n(v)$ while not allowing the entire distribution to be cooled and slowed to the zero crossing of $F(v)$.[8] The probe was focused to ~ 300 μm in diameter and aligned so as to overlap the full diameter of the atomic beam at the 17-cm observation point. The probe fluorescence was observed in a direction perpendicular to both the probe beam and the standing wave. To make the measurements, the standing-wave field was turned off for 16 μsec during which time the probe field was pulsed on. The optical switching was performed using an acousto-optic modulator. The probe field pulses were ~ 10 μsec in duration at a repetition rate of ~ 3 Hz. Thus $n(v)$ was determined by measuring the fluorescence due to the probe as a function of probe frequency with the standing-wave frequency held constant. The angle between the probe and the atomic beam was 2 mrad, yielding a longitudinal velocity resolution of ~ 6 m/sec, due to the resolution limit of the natural linewidth Γ for the Doppler fluorescence technique used. The measured $n(v)$ was shown to be independent of the intensity of the weak probe ($I \sim 20$ μW/cm^2 $\ll I_{sat}$). Indeed, in the absence of the standing wave, the velocity distribution of the atomic beam was well characterized by a Maxwell-Boltzmann distribution at a temperature $T \approx 180\,^\circ$C, in good agreement with a thermometer connected to the oven.

A weak magnetic field (~ 5 G) along the axis of the SW and the atomic beam defined a quantization axis for the atoms; therefore, for circularly polarized light in the SW field, the atoms are optically pumped into the $m_F = 2$ sublevel of the $F = 2$ ground state. The atomic population in the $F = 2$ ground state [i.e., $n(v)$] was shown to be almost exclusively confined to the $m_F = 2$ ground-state sublevel by measuring the probe fluorescence as a function of probe polarization. Thus the atom interacting with the field is well approximated by a two-level model.

The approximate position, size, and width of the Doppleron resonances can be understood considering a simple physical picture. The resonances result from processes in which an atom absorbs $N+1$ photons from one TW and is *stimulated* to reemit N photons into the oppositely directed TW. The final photon is *spontaneously* emitted. This point is essential to understanding why the Doppleron resonances can give rise to such large accelerations. For $\Delta < 0$, the atom absorbs the $N+1$ photons from the counterpropagating traveling wave, which has detuning $\Delta + |kv|$ in the atom's rest frame. Similarly, the copropagating field will have frequency $\Delta - |kv|$ in the same rest frame. The total-energy change will be

$$\hbar[(\omega+|\mathbf{k}\cdot\mathbf{v}|)(N+1)-(\omega-|\mathbf{k}\cdot\mathbf{v}|)N-\omega_0] = \hbar[\Delta+(2N+1)|\mathbf{k}\cdot\mathbf{v}|]\,,$$

where $\omega = \omega_0 + \Delta$. The resonance condition is

$$|\mathbf{k}\cdot\mathbf{v}| = -\Delta/(2N+1)\,. \qquad (1)$$

The lowest-order process occurs for $N=0$ and $|\mathbf{k}\cdot\mathbf{v}| = -\Delta$, which is simply the position of the spontaneous force peak for a single counterpropagating TW. Since each photon carries a momentum $\hbar\mathbf{k}$, and the net average momentum transfer associated with spontaneous emissions is zero, the net average momentum transfer associated with this process is $\hbar\mathbf{k}$. The characteristic rate for this process is $\Gamma/2$ and hence the maximum force is $\hbar\mathbf{k}\Gamma/2$. The Doppler width of the resonance (in velocity) will be of the order of the power-broadened linewidth. The next highest Doppleron resonance, $N=1$, occurs at $|\mathbf{k}\cdot\mathbf{v}| = -\Delta/3$, and imparts a net average momentum of $3\hbar\mathbf{k}$ to the atom. However, the rate is still limited by the single-spontaneous-emission rate $\Gamma/2$, and so the maximum force $\approx 3\hbar\mathbf{k}\Gamma/2$ is greater than 3 times the maximum single-photon spontaneous force $3\hbar\mathbf{k}\Gamma/6$. Notice that the increase in the average longitudinal force (the average force along the SW) does *not* involve an increase in the transverse diffusion rate (diffusion rate perpendicular to the atomic beam) because only a single spontaneous emission is involved. Since the $N=1$ process is a three-photon process, the width of the $N=1$ resonance is narrower than the width of the one-photon $N=0$ process. This argument extends to arbitrarily large N, where the width of the resonances in velocity space can be much narrower than the Doppler spread of the natural linewidth (see Fig. 2). For a given I and Δ the force associated with the Nth Doppleron is proportional to the maximum possible force $[(2N+1)\hbar\mathbf{k}\Gamma/2]$ weighted by the probability for the $(2N+1)$-photon process. This probability is a decreasing function of N, since the total number of photons in the field is limited. Thus the amplitude of the Doppleron peaks must eventually decrease at very large N. In sum then, the Doppleron resonances represent a coherent redistribution of photons between the two TWs which form the SW. As a result, $a(v)$ displays a series of resonances at $|kv| = -\Delta/(2N+1)$, where the $(N+1)$th resonance is narrower than the Nth. For moderate I and small N the amplitude of successive resonances increases with N, but will eventually decrease to zero at large N.

The true position of the Doppleron peaks deviates from Eq. (1), particularly at high intensities ($I \gg I_{sat}$). This discrepancy arises primarily because the simple argument above neglects the Stark shift associated with the light field itself. The effective Stark shift of one TW component of the SW with respect to the *other* TW component can give rise to shifts in the effective two-level resonant frequency.[10] The latter effect is valid for small N, and hence larger v, when the separation of the force due to the SW into a sum of forces due to the two counterpropagating TWs is most accurate. At low velocities, and hence near higher-N Dopplerons, such a separation can no longer be made and, in fact, the Doppleron positions and their variations with Δ are more closely predicted by the simple model. If I is increased, the Stark shift will increase, and the positions of the Doppleron peaks will move to higher velocities. In addition, as I increases the value of N for which the peak amplitude begins to decrease will also increase (see above). As I was decreased, a decrease in the velocities at which the peaks in $n(v)$ occur was experimentally observed, consistent with this discussion.

We have performed numerical simulations which model the evolution of a Maxwell-Boltzmann velocity distribution under the influence of the optical-standing-wave force. The force was calculated using a continued-fraction solution.[4] We note that in the continued-fraction analysis the Doppleron resonances correspond to successive zeros in the numerators of the convergent-fraction series. Using this approach, we were able to allow both for deviations from Eq. (1) described above and for the finite atom-field interaction length used in our experiments. A sample is shown by the dashed line in Fig. 2. In this simulation, only the longitudinal force was considered and the SW was treated as an infinite plane wave. When the finite transverse extent of the SW was included in the calculation, little improvement was seen in the fits. These observations suggest that the measured $n(v)$ is dominated by atoms which do not spend significant time in the low-intensity regions of the SW beam. We note that a non-plane-wave beam profile can couple the strong slowing and cooling effects of the longitudinal Doppleron resonances to the transverse motion. This coupling may be responsible for significant changes in the transverse distribution of the atoms. In our simulations, we have also neglected the effects of transverse guiding,[9] which may contribute to the difference in peak heights between the data and the fit.

By studying the variation of the Doppleron-induced holes in $n(v)$ as a function of Δ we were able to assign an

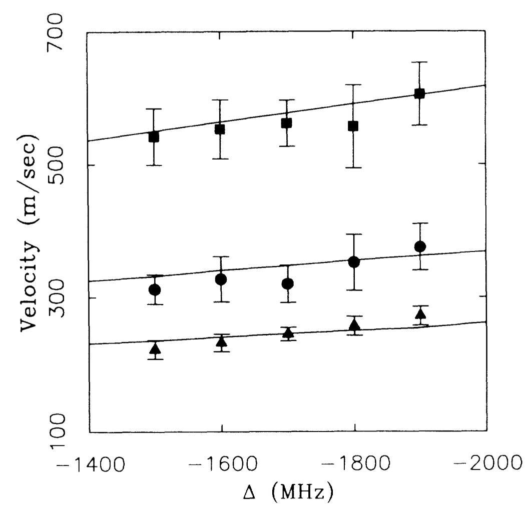

FIG. 3. Position of Doppleron resonances in $n(v)$ vs Δ. From the top down, the resonances are labeled by $N=0$, 1, and 2 (see text). The solid lines are the positions of the Dopplerons as determined from the theoretical $a(v)$ curves (see bottom graph in Fig. 2). Horizontal error bars are smaller than symbol size.

index number N to the observable resonances (see Fig. 3). The data shown are restricted to the range in Δ where the given minima in $n(v)$ associated with a particular Doppleron were well defined and could easily be followed with variations in Δ. The solid lines correspond to the theoretical position for the first three Doppleron resonances as determined by the force curves used in the simulations. Although the data clearly cluster about the theoretical curves, the expected dependence of Doppleron position on Δ is not well determined over the limited frequency range.

Perhaps the most important aspect of these observations is the extremely rapid changes in acceleration as a function of velocity which occur as a result of the Doppleron resonances. In fact, the Dopplerons give rise to rapid changes in all of the internal coordinates of the atom (i.e., the Bloch vectors) as a function of velocity. One example of this is the sharpness of the Doppleron resonances as seen in the force-versus-velocity curve. For the conditions of these experiments, changes in atomic decelerations in excess of 5×10^6 m/sec^2 over a velocity range smaller than the natural linewidth (~6 m/sec) are predicted by the theory (see lower part of Fig. 2). For an acceleration of 5×10^6 m/sec^2, the change in velocity over a few spontaneous emission times τ_{sp} is sufficient to drastically alter the force experienced by the atom. In the continued-fraction solution for the force, the atomic velocity is assumed to be constant over τ_{sp}. This is the so-called adiabatic approximation. We point out that in light of the large decelerations associated with the Doppleron resonances, the rate of change in velocity with respect to the rate of change of the internal coordinates of the atom are not negligible; consequently, the adiabatic approximation is no longer valid.[4] The extent to which the atomic motion differs from the predictions of the continued-fraction calculation is not easily extracted from our data.

It is our hope that this prospect will stimulate renewed theoretical interest in the motion of atoms in intense standing-wave fields, and that indeed additional new phenomena may be predicted and observed.

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