Recoil-Induced Resonances in Cesium: An Atomic Analog to the Free-Electron Laser

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We report on the first experimental observation of recoil-induced resonances. These resonances, which can be considered as the atomic analog of the gain mechanism of the free-electron laser, are observed on the transmission spectrum of a probe beam making a small angle with a one-dimensional lin⊥lin cesium optical molasses. They are interpreted either as stimulated Raman transitions between differently populated velocity groups (the internal atomic state being unchanged), or as stimulated Rayleigh resonances originating from a bunching of the atomic density. Their shape provides information about the transverse velocity distribution of the atoms.

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The possibility of converting the kinetic energy of free electrons into radiation energy is well known and has led to the development of the free-electron laser [1]. The gain mechanism of such a laser originates from energy transfer between two counterpropagating electromagnetic fields interacting with an ensemble of free electrons at thermal equilibrium [2]. Two different, though equivalent, physical interpretations of the gain process are found in the literature. The first one [2] is based on the quantization of the electron translational degrees of freedom in *momentum space*. The laser action is then interpreted in terms of stimulated Compton scattering. In the second interpretation [3], the electron translational degrees of freedom are described in *position space* and the amplification process arises from a stimulated Rayleigh scattering produced by a bunching of the electron density. The possibility of observing an analogous gain mechanism in the case of an atomic vapor was considered theoretically by Guo *et al.* [4], who showed that it should lead to a new kind of resonance in nonlinear spectroscopy (called recoil-induced resonance). In this Letter, we report on what is, to our knowledge, the first unambiguous experimental observation of these resonances. They are observed on the transmission spectrum of a weak probe beam interacting with a sample of laser-cooled cesium atoms in the one-dimensional lin⊥lin geometry [5]. By analogy with the free-electron laser, we show that the recoil-induced resonances can be interpreted either in terms of stimulated *Raman* scattering between differently populated atomic velocity groups [4], or in terms of stimulated *Rayleigh* scattering from a bunching of the atomic density. Finally, we show that their shape provides information about the atomic velocity distribution.

Consider an ensemble of two-level atoms interacting with a pump beam of frequency ω and wave vector $\mathbf{k}=k\mathbf{e}_z$, and a probe beam having the same polarization, frequency $\omega_p=\omega+\delta$, and wave vector $\mathbf{k}_p=k_p\cos\theta\mathbf{e}_z+k_p\sin\theta\mathbf{e}_y$ [Fig. 1]. The central resonance of the probe transmission spectrum results from two competing processes: on one hand, a gain mechanism provided by stimulated photon redistribution from the pump to the probe beam associated with a change in atomic momentum from $\mathbf{p}$ to $\mathbf{p}+\hbar(\mathbf{k}-\mathbf{k}_p)$ (because of momentum conservation); on the other hand, an absorption mechanism associated with the reverse process with modification of the atomic momentum from $\mathbf{p}+\hbar(\mathbf{k}-\mathbf{k}_p)$ to $\mathbf{p}$. However, *because the populations* $\pi(\mathbf{p})$ *and* $\pi[\mathbf{p}+\hbar(\mathbf{k}-\mathbf{k}_p)]$ *of the initial and final velocity groups of the scattering process are generally different*, one redistribution process overcomes the other, and the probe beam eventually gains (or loses) energy, proportionally to the population difference $\pi(\mathbf{p})-\pi[\mathbf{p}+\hbar(\mathbf{k}-\mathbf{k}_p)]$. The recoil-induced resonance results from these stimulated Raman scattering processes, averaged over the total atomic momentum distribution. In the simple case where the angle θ between the pump and the probe beams is sufficiently small that terms of the order of θ^2 can be neglected, it is straightfor-

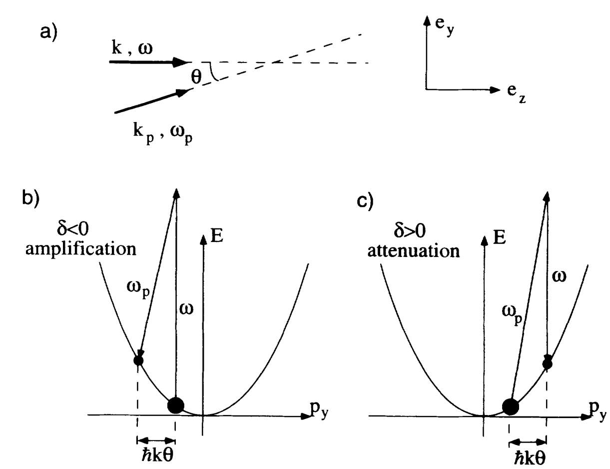

FIG. 1. Recoil-induced resonances. (a) Scheme of the fields. (b) and (c) Stimulated Raman transitions between free atomic external states having energies $E=p_y^2/2M$. Because the population of the velocity groups is a decreasing function of $|p_y|$, there is absorption of the high-frequency field and amplification of the low-frequency field.

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ward to derive the line shape of this resonance. Using energy and momentum conservation laws, one finds that the elementary contribution of a given velocity group $\mathbf{p}$ corresponds to a Lorentzian Raman resonance centered around $\delta = k\theta p_y/M$ (where M is the atomic mass) and of homogeneous width 2γ (γ is a typical population relaxation rate). As a result, the amplification coefficient of the probe field is proportional to [4]

$$g(\delta) = \frac{\Omega^2\Omega_p}{\Delta^2}\int dp_y \frac{\gamma[\pi(p_y) - \pi(p_y - \hbar k\theta)]}{\gamma^2 + (\delta - k\theta p_y/M)^2}, \quad (1)$$

where Ω and Ω_p are, respectively, the resonance Rabi frequencies for the pump and probe fields, and Δ is the frequency detuning from resonance (assumed to be much larger than the natural width of the excited state). The term in front of the integral corresponds to the usual Raman coupling, proportional to the pump intensity times the probe amplitude, and inversely proportional to the square of the detuning. The integral over all atomic momenta results from the fact that the levels involved in the stimulated Raman processes belong to a *continuum.* If $\pi(p_y)$ has a slow variation in the interval $\hbar k\theta$, one can substitute $\hbar k\theta \partial\pi(p_y)/\partial p_y$ for $\pi(p_y) - \pi(p_y - \hbar k\theta)$ into Eq. (1). By assuming a Maxwell-Boltzmann distribution for the atomic momentum having a rms momentum $M\bar{v}$, one then finds that the shape of the recoil-induced resonance is a convolution product of a Lorentzian of width $2M\gamma/k\theta$ and of the derivative of a Gaussian of width $2M\bar{v}$. We restrict ourselves to the limit of small relaxation rate $\gamma \ll k\theta\bar{v}$ which is equivalent to the large cavity limit in the free-electron laser, yielding large amplification factors [2,3]. It allows us to substitute a Dirac delta function for the Lorentzian in Eq. (1), which then reads

$$g(\delta) = -\left(\frac{\pi}{2}\right)^{1/2} \frac{\hbar\Omega^2\Omega_p}{\Delta^2 k_B T}\frac{\delta}{k\theta\bar{v}} \exp\left[-\frac{\delta^2}{2(k\theta\bar{v})^2}\right], \quad (2)$$

where k_B is the Boltzmann constant and T is the temperature of the atomic medium ($k_B T = M\bar{v}^2$). The recoil-induced resonance thus appears as a structure centered in $\delta = 0$ with a Gaussian derivative shape [4]. One notices that the probe field experiences amplification when $\delta < 0$ and attenuation when $\delta > 0$, whatever the sign of the detuning Δ. The width $2k\theta\bar{v}$ of the recoil-induced resonance is proportional to the rms atomic velocity, and thus gives *in situ* access to the atomic temperature. Moreover, the primitive of the resonance profile exactly corresponds to the atomic momentum distribution [6]. The amplitude of the recoil-induced resonance is independent of the angle θ as long as $\gamma \ll k\theta\bar{v}$. Consequently, these resonances can persist at the center of the probe transmission spectrum even for values of θ as small as 10^{-2} rad. A small but *nonzero* angle between the probe and the pump beams can thus yield a probe transmission spectrum qualitatively different from the one expected at zero angle [7].

We now present an interpretation of the recoil-induced resonances in classical terms by describing the atomic system in position space. Combining the probe and the pump fields, one obtains a wave whose intensity exhibits a time and spatially modulated part (frequency δ, wave vector $\mathbf{k}_p - \mathbf{k} \approx \theta k\mathbf{e}_y$). This component leads to a transverse spatial modulation of the light shifts of the atomic internal states, which acts as a potential of depth $U_p = \hbar\Omega\Omega_p/2|\Delta|$ for the atomic motion, and hence to a spatial bunching of the atomic density in the transverse direction. The subsequent diffraction of the pump beam from this atomic density grating leads to a wave having the same frequency and direction of propagation as the probe beam. Because of the finite response time of the medium ($\approx 1/\gamma$), the spatial modulation of the atomic density is not in phase with the modulation of the light shifts, so that the diffracted beam has a component in phase with the probe, and can interfere with it [8]. Following Ref. [3], one can recover the shape of the recoil-induced resonance in a quantitative way, by deriving the atomic density in phase space from the classical Boltzmann equation. Thus, two different though equivalent physical interpretations can be given, depending on whether the derivation is carried in momentum or position space. Recoil-induced resonances therefore provide a new illustration of the counterintuitive duality between stimulated Raman and Rayleigh scattering. Apart from its intrinsic interest, this duality permits a more profound understanding of these resonances. First, the dependence of their amplitude on the atomic temperature becomes clear when noticing that the Raman coupling term in front of the integral (2) corresponds to the ratio $U_p/k_B T$, which can be considered as a Boltzmann-type factor. Second, the condition $\gamma \ll k\theta\bar{v}$ for obtaining large resonance amplitude can be reinterpreted since the ratio $k\theta\bar{v}/\gamma$ means that an atom typically experiences several periods of the optical potential before its free motion is modified significantly.

We now present the experimental setup that was used to observe the recoil-induced resonances. The previously discussed conditions required for the observation of these resonances impose severe constraints on the experiment. First, because the resonance amplitude is inversely proportional to the atomic temperature [see Eq. (2)], we used a laser-cooled sample of cesium atoms. However, because such samples have low densities, the incoming beams have to be nearly resonant. As a result, in an experiment performed in the geometry of Fig. 1(a), the atoms can be expelled from the interaction regin by radiation pressure. To prevent this effect, the pump beam was reflected onto itself, but its linear polarization was rotated by 90°. We thus obtained in a transient way a one-dimensional lin$\perp$lin optical molasses [5], starting from atoms cooled and loaded in a magneto-optical trap [9]. In this geometry, however, additional Rayleigh resonance (associated with the longitudinal atomic motion) is expected at the center of the probe transmission spectrum [10,11], which can prevent the observation of recoil-induced resonances (associated with the transverse atomic motion). In fact, it was shown recently [11,12] that for a probe beam having the same linear polarization as the copropagating pump beam [Fig. 1(b)], these additional resonances have generally an amplitude much smaller than the recoil-induced resonances and opposite sign [13]. Second, because the recoil-induced resonances are associated with a *free* atomic motion in the transverse direction, the residual trapping fields that were used in [5] to damp this motion and hence increase the lifetime of the molasses were here *completely switched off* (we have indeed observed that the shape of the central resonance changes dramatically in the presence of a weak residual trapping field).

The central part of the probe transmission spectrum is shown in Fig. 2 for different values of the frequency detuning Δ and a pump intensity of 50 mW/cm^2. It displays a resonant structure having a width of the order of 10-20 kHz and an amplitude corresponding to a probe gain of $\approx 10^{-2}$. In each case, the central resonance has

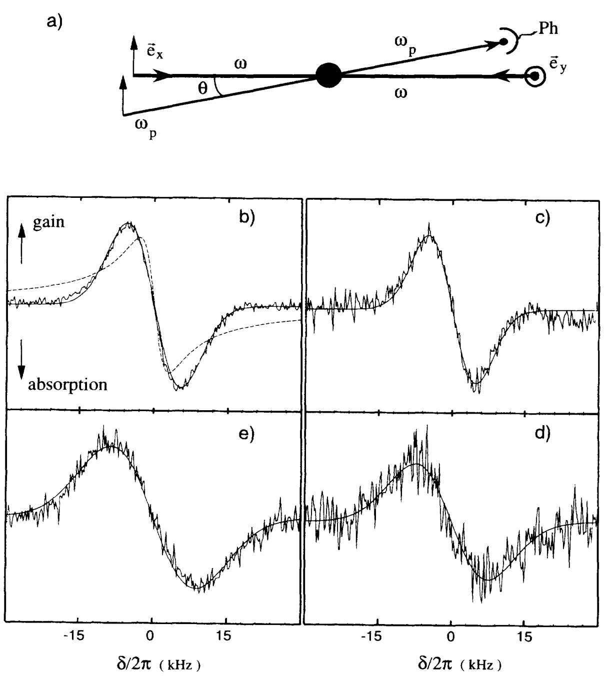

FIG. 2. (a) Scheme of the fields in the experiment. Central resonance of the probe transmission spectrum versus $\delta = \omega_p - \omega$ (sweeping rate: 60 kHz/ms) for $\Delta/\Gamma = -10$ (b), -22 (c), and $+7$ (d). The intensities of the pump and the probe beams are, respectively, 50 mW/cm^2 and 0.1 mW/cm^2, and $\theta \approx 2.3°$. The fit with the derivative of a Gaussian is shown in solid lines. We show in (b) the best fit with a dispersion (dashed line), which corresponds to the usual shape of central resonances in nonlinear spectroscopy. The comparison clearly shows that the derivative of a Gaussian is in better agreement with experimental observations. The curve in (e) is obtained in the same conditions as (b), but for the fact that the probe is applied 1 ms after the pump beams. The difference in width results from transverse heating.

a shape which shows a very good fit with a derivative of a Gaussian. Furthermore, probe gain is always observed for $\delta \leq 0$, *whatever the sign of the frequency detuning.* These features were observed in the whole range of parameters $5\Gamma \leq |\Delta| \leq 30\Gamma$ and 5 $mW/cm^2 \leq I \leq 50$ mW/cm^2 that we have studied, and are consistent with recoil-induced resonances [14]. We performed different tests of the proportionality between the width of the resonance and the Doppler width of the atomic medium $k\theta\bar{v}$. First, the atomic temperature deduced from the resonance widths of Figs. 2(b) and 2(c) is found to be equal to ≈ 0.1 mK, a typical temperature in a magneto-optical trap. In addition, when changing the temperature of the cloud of trapped atoms, the resonance width was observed to vary accordingly. Second, in the interval $2° \leq \theta \leq 6°$, the resonance width was observed to vary linearly with θ, as expected. Third, probe transmission spectra were recorded at different times after the trapping beams had been switched off. In this case, transverse heating occurs because of absorption-spontaneous emission cycles, which increase the rms velocity $\bar{v}$ along the y axis and lead to a broader central resonance, as was observed experimentally [Figs. 2(b) and 2(e)]. Another piece of information can be deduced from the widths of the curves in Fig. 2. Because these widths are typically of the order of 10 kHz, and because the resonances have the shape of a Gaussian derivative, one deduces that the *homogeneous width γ is smaller than $2\pi \times 10^4$ s^{-1} and thus is much smaller than the total scattering rate* Γ' (which is of the order of 10^6 s^{-1}). This feature *cannot be accounted for by any localization effect* leading to a Lamb-Dicke narrowing of the resonance as in [5,10], because atomic motion is free along the transverse direction. The occurrence of long evolution times can here be associated with the atomic momentum distribution's being much larger than $\hbar k$. As a result, the distribution of populations and coherences between external states having neighboring momenta does not change very much after an absorption-spontaneous emission cycle; hence the relaxation of these quantities involves many scattering processes.

In the case of two-dimensional (2D) molasses, a similar recoil-induced resonance is expected when the probe beam does not propagate in the plane of the molasses beams. In the case of 3D optical molasses or lattices (as well as 2D molasses when the probe and the molasses beams propagate in the same plane), it seems more appropriate to describe the central resonance in terms of stimulated Rayleigh scattering, by using a 3D generalization of the approach described in Ref. [10]. However, the contribution to the central resonance of the unbound atoms can exhibit properties analogous to recoil-induced resonances [15].

The interest of the recoil-induced resonances goes far beyond the interpretation of pump-probe spectra of optical molasses. It is a powerful tool for characterizing the momentum distribution of a system which may prove interesting as a substitute for the time of flight technique for temperature measurements in optical molasses. In fact, its range of application is broader than laser-cooled atomic samples. It might be used to probe the momentum distribution of atoms of molecules cooled by cryogenic techniques, even by using very off-resonant lasers. For example, using the green line of an Ar^+ laser in a cavity of Q factor 100 and cold polarized hydrogen [16], one expects a probe gain of the order of 10^{-6}–10^{-4}, which is small but still seems measurable.

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[6] This result holds for any momentum distribution, provided that $\gamma \ll k\theta\bar{v}$.

[7] It can be shown that in the limit where $\gamma \gg k\theta\bar{v}$, the recoil-induced resonance appears as the derivative of a Lorentzian of width 2γ whose amplitude, proportional to $(k\theta\bar{v}/\gamma)^2$, vanishes as θ tends toward zero.

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[12] Since this paper was submitted for publication, we received a preprint of J. Guo, who shows theoretically that the recoil-induced resonances are 1 order of magnitude larger than the longitudinal Rayleigh resonances in the geometry of our experiment.

[13] These properties also indicate that the central resonance shown in Fig. 1(c) of Ref. [5] was probably dominated by recoil-induced effects.

[14] It may be noticed that these features were *not* observed in one-dimensional *corkscrew* optical molasses [B. Lounis *et al.*, Phys. Rev. Lett. **69**, 3029 (1992)]. In that case, the longitudinal Rayleigh resonance, which results from a very different physical process [J.-Y. Courtois and G. Grynberg, Phys. Rev. A **48**, 1378 (1993)], has a larger amplitude than the recoil-induced resonance.

[15] An earlier experiment showed that the width of the central resonance in a 3D σ^+-σ^- molasses varies linearly with the angle between the probe and the closest molasses beam [B. Lounis *et al.*, Nonlinear Opt. **5**, 459 (1993)]. This observation may reflect a contribution of recoil-induced effects.

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