Radiation Pressure in a Rubidium Optical Lattice: An Atomic Analog to the Photorefractive Effect

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Probe gain in a rubidium optical lattice is observed when the probe and lattice beams have identical frequencies. This effect is shown to arise from the radiation pressure that shifts the atomic density distribution with respect to the optical potential. This effect is compared with two-beam coupling in photorefractive materials. The experimental results obtained by changing the parameters of the optical lattice (intensity, detuning, periodicity) are in reasonable agreement with numerical simulations based on the model case of a $1/2 \rightarrow 3/2$ atomic transition. [S0031-9007(97)03427-3]

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Two-beam coupling [1] between *frequency-degenerate* laser beams is one of the most intriguing phenomena in nonlinear optics. Owing to the apparent symmetry of the interaction between two such beams and a nonlinear medium, one indeed expects no net power transfer to take place between the laser waves. Of course, the symmetry can be broken through the intensity, direction, or polarization of the beams. Still, the possibility of observing an actual amplification of one wave at the expense of the other is a rather uncommon phenomenon. This can be better understood by identifying the conditions required in such a process. The intensity attenuation or amplification of a laser beam during propagation through a material medium arises from the existence of a macroscopic polarization (i) having the same characteristics (frequency and wave vector) as the incident beam, (ii) but having a $\pi/2$ phase-shifted component with respect to this beam. In the more particular case of two-beam coupling where the intensity modification only occurs in the presence of a supplementary beam, the polarization of interest is necessarily nonlinear in nature. Although nonlinear interactions naturally provide polarization components fulfilling requirement (i) through the creation of gratings in the material medium by the interference pattern between the incident waves, condition (ii) is very seldom fulfilled because, most generally, a stationary interference pattern (the incident laser beams have the same frequency) can only induce 0 or π phase-shifted steady-state material gratings.

The photorefractive effect [2] taking place in some particular crystals such as $LiNbO_3$ or $BaTiO_3$ is one (if not the only) outstanding counterexample of this intuitive general property. In such crystals, the spatially modulated intensity distribution $I(\mathbf{r})$ due to the interference between the two incident beams generates charge carriers in the conduction band through photoexcitation, at a rate proportional to the local value of the optical intensity. Because of spatial diffusion through the crystal, the carriers migrate towards points of smaller intensity where they recombine with donors. This results in a charge density $\rho(\mathbf{r})$ exhibiting a π phase shift with respect to $I(\mathbf{r})$. Because of the Poisson equation $[\nabla \cdot (\varepsilon \mathbf{E}) = \rho]$, the spatially varying charge distribution $\rho(\mathbf{r})$ gives rise to a nonuniform electric field $E(\mathbf{r})$, which is phase shifted $\pi/2$ with respect to $I(\mathbf{r})$. Finally, $E(\mathbf{r})$ yields a proportional refractive index grating $n(\mathbf{r})$ through the linear electro-optic effect (Pockels effect). Thus, by transforming an incident interference pattern $I(\mathbf{r})$ into a $\pi/2$ phase-shifted grating $n(\mathbf{r})$, the photorefractive effect allows power exchange between frequency-degenerate laser beams [3].

In this Letter, we present and experimentally demonstrate an analog of this seemingly very peculiar effect in the case of an atomic gaseous medium. More precisely, we show that the transmission spectrum of a weak probe beam interacting with a standard three-dimensional lin $\perp$ lin optical lattice [4] can display a narrow *Lorentzian-like* central resonance. The probe gain in the frequency-degenerate regime can reach up to 10% and can be interpreted as a two-beam coupling process involving the creation of an atomic density (or equivalently a refractive index) grating phase shifted $\pi/2$ with respect to the stationary probe-lattice wave. We show that this phenomenon arises from the *radiation pressure* exerted by the probe-lattice pattern onto the atoms. This proves that the dissipative forces, although generally neglected, can have a dramatic effect on the spatial distribution of atoms in optical lattices. Furthermore, we show that the width of the central resonance is related to the spatial diffusion tensor [5] of the atoms in the lattice, which can thus be investigated directly.

Standard four-beam optical lattices [4] consisting of two x-polarized beams propagating in the yOz plane and making an angle $2\theta_y$, and two y-polarized beams propagating in the xOz plane and making an angle $2\theta_x$ [see Fig. 1(a)], have been intensively investigated during the last few years. Many properties of this system have been already revealed: oscillating motion of the atoms in the optical potential wells associated with the light shifts [6,7], narrowing of the vibrational lines due to strong spatial confinement at the bottom of the wells [8], paramagnetic behavior [9], long range spatial order [10], propagating elementary excitation modes [11], etc. Curiously

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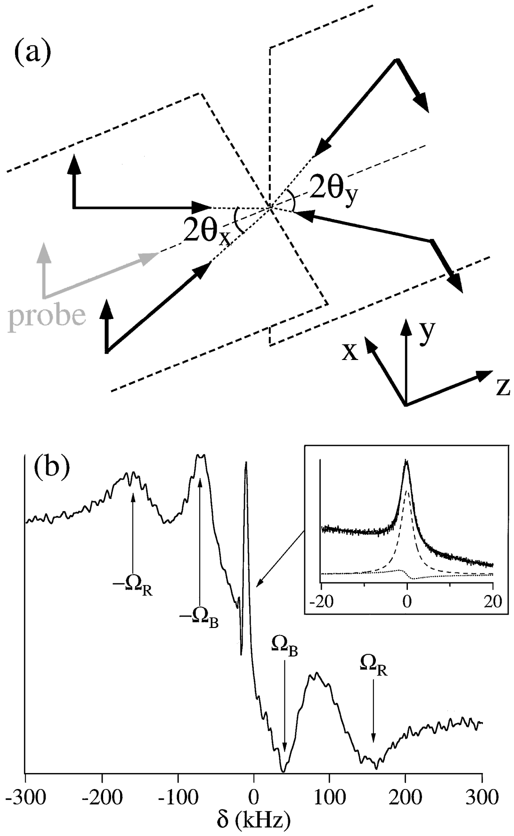

FIG. 1. Probe transmission spectroscopy in a 3D lin $\perp$ lin rubidium lattice. (a) Interaction geometry. The probe beam propagates along the symmetry axis Oz of the lattice, making an angle θ_x with the nearly copropagating lattice beams. (b) Probe transmission spectrum recorded for $\theta_x = \theta_y = 20°$, a lattice beams intensity $I = 5\ \mathrm{mW\,cm^{-2}}$, and a frequency detuning $\Delta = -5\,\Gamma$. One clearly distinguishes Raman (Ω_R) and Brillouin-like (Ω_B) resonances, together with a remarkably narrow central resonance structure displayed in the inset. The fit using an adjustable superposition of a Lorentzian and a dispersion having the same width shown in the inset proves that the resonance is very close to a Lorentzian. This resonance has a HWHM of 1.4 kHz and corresponds to a probe amplification of 5%.

enough, no systematic study of the central part of probe transmission spectra in a four-beam optical lattice has been reported so far, although it is known to provide unique information about transport properties of the atoms [8,12]. It turns out that in the limit of small θ_x and θ_y angles and in the situation where the probe beam propagates along the symmetry axis, Oz, with the same polarization as the nearly copropagating lattice fields, the central feature of the spectra displays outstanding characteristics. We show in Fig. 1(b) a transmission spectrum recorded following the experimental procedure described in [6] in a rubidium lattice with $\theta_x = \theta_y = 20°$, a lattice beams intensity $I = 5\ \mathrm{mW\,cm^{-2}}$, and a red frequency detuning $\Delta = -5\Gamma$ between the lattice beams and the $5S_{1/2}F = 2 \rightarrow 5P_{3/2}F' = 3$ resonance frequency of ^{87}Rb (Γ: natural linewidth of the excited state). One clearly distinguishes the previously reported Raman [6,7] and Brillouin-like [11] resonances, together with a narrow central structure having a very unusual shape. A magnification of this resonance is displayed in the inset of Fig. 1(b), where it appears as an almost pure Lorentzian structure having a HWHM of 1.4 kHz superimposed on a background slope originating from a much broader dispersivelike resonance. Very surprisingly, the amplification of the probe is maximum when its frequency equals that of the lattice beams, in contrast with the dispersivelike stimulated Rayleigh resonances previously reported in optical lattices [6,8], corresponding to zero probe gain in the frequency-degenerate regime. Where does such an unusual observation come from?

Optical pumping, as well as radiation pressure, involves photon absorption and is thus proportional to the light intensity. The dipole potential arises from the interaction of the laser electric field with the laser-induced atomic dipole and is also proportional to the light intensity. The dipole force, which derives from the dipole potential, is therefore phase shifted $\pi/2$ with respect to the laser intensity, hence to the radiation pressure force. Because when neglecting radiation pressure, the atomic density has extrema at points where the dipole force vanishes, it turns out that *radiation pressure* is the *only* possible mechanism leading to a phase shift of the atomic density distribution with respect to a *stationary* pump-probe intensity modulation, hence for the nonzero probe gain at the center of the spectrum.

One can wonder why these radiation-pressure induced central resonances have not been discussed previously. It is usually argued that because the radiation pressure force scales as $I\Gamma/\Delta^2$, whereas the dipole force scales as I/Δ, in the usual experimental conditions where $\Delta \gg \Gamma$, radiation pressure is negligible compared to the dipole force. In fact, this argument is only partially correct as it omits the influence of the laser interaction geometry. Consider a pump and a probe beam having wave vectors $\mathbf{k}$ and $\mathbf{k}_p$ and frequencies ω and $\omega_p = \omega + \delta$, respectively. The modulated part of the light intensity arising from their interference takes the form $I(\mathbf{r}) \propto \sqrt{II_p}\cos[\delta t - (\mathbf{k}_p - \mathbf{k})\cdot\mathbf{r}]$. As a result, the modulated part of the dipole force reads $\mathbf{F}_d(\mathbf{r}) \propto [\sqrt{II_p}/\Delta](\mathbf{k}_p - \mathbf{k})\sin[\delta t - (\mathbf{k}_p - \mathbf{k})\cdot\mathbf{r}]$, where the $\mathbf{k}_p - \mathbf{k}$ factor is reminiscent of the dipole force being due to photon redistribution processes between the pump and probe waves, hence a wave-vector difference. By contrast, the modulated part of the radiation pressure force takes the form $\mathbf{F}_r(\mathbf{r}) \propto [\sqrt{II_p}\,\Gamma/\Delta^2](\mathbf{k}_p + \mathbf{k})\cos[\delta t - (\mathbf{k}_p - \mathbf{k})\cdot\mathbf{r}]$, where the $\mathbf{k}_p + \mathbf{k}$ factor accounts for an *identical* enhancement or reduction in the number of photons absorbed in each of the two waves, hence a sum of wave vectors. The actual condition for the radiation pressure *not* to be negligible compared to the dipole force is thus $\Delta/\Gamma \leq |\mathbf{k}_p + \mathbf{k}|/|\mathbf{k}_p - \mathbf{k}|$ which is much less stringent than $\Delta/\Gamma \leq 1$ when $|\mathbf{k}_p - \mathbf{k}|$ happens to be small, i.e., when the pump-probe angle is small.

It is also legitimate to wonder what the width of the central resonance corresponds to. In order to answer this question, we will make use of a phenomenological description of the atomic dynamics in the lattice. We consider a two-dimensional model where the atoms propagate in the x-z plane connecting the potential minima. The atomic density distribution $P(\mathbf{r}, t)$ is supposed to obey the following Fokker-Planck equation:

$$\partial_t P + \nabla \cdot \mathbf{J} = 0, \qquad (1)$$

$$J_i = -D_i[\partial_i P - P(F_{d,i} + F_{r,i})/k_B T]. \qquad (2)$$

In Eq. (2), J_i, $F_{d,i}$, and $F_{r,i}$ denote the i component of the flux and of the dipole and radiation pressure forces experienced by the atoms in the presence of the lattice and probe fields, D_i is the spatial diffusion coefficient of the atoms in the lattice along the i direction, k_B is the Boltzmann constant, and T is the lattice temperature [13]. We solve Eq. (1) by using perturbation theory, with the ratio ϵ between the probe and lattice field amplitudes as the small parameter. Assuming $\theta_x = \theta_y = \theta$ so that the mean value of $\mathbf{F}_r$ cancels at zeroth order in ϵ and denoting by $U^{(0)}(\mathbf{r})$ the lowest optical potential due to the lattice field alone, the stationary solution of (1) in the absence of the probe is found to take the well-known form $P^{(0)}(\mathbf{r}) \propto \exp[-U^{(0)}(\mathbf{r})/k_B T]$. To first order in ϵ, the dipole and radiation pressure forces are modified in the form

$$\mathbf{F}^{(1)}(\mathbf{r}, t) = \sum_{i=1,2} \mathbf{f}_i^{(1)} \exp[-i\delta t + i(\mathbf{k}_p - \mathbf{k}_i) \cdot \mathbf{r}] + \text{c.c.}$$
$$\equiv \sum_{i=1,2} \mathbf{f}_i^{(1)} \exp[-i\delta t + i\mathbf{K}_i \cdot \mathbf{r}] + \text{c.c.}, \qquad (3)$$

where $\mathbf{K}_i$ is the difference between the probe ($\mathbf{k}_p$) and either of the nearly colinear lattice wave vectors ($\mathbf{k}_1$ and $\mathbf{k}_2$), which contribute independently to $\mathbf{F}^{(1)}$. Note that because of the previously mentioned properties of $\mathbf{F}_d$ and $\mathbf{F}_r$, $\mathbf{f}_{d,i}^{(1)}$ is purely imaginary and has components along x and z proportional to $K_{i,x}$ and $K_{i,z}$, respectively, whereas $\mathbf{f}_{r,i}^{(1)}$ is purely real and its dominant component is aligned along Oz. Equation (1) can then be conveniently solved by writing $P^{(1)}$ in the form

$$P^{(1)}(\mathbf{r}, t) = \sum_{i=1,2} P^{(0)}(\mathbf{r}) a_i^{(1)} \exp[-i\delta t + i\mathbf{K}_i \cdot \mathbf{r}] + \text{c.c.},$$

which yields after a straightforward calculation [14]

$$a_i^{(1)} = \frac{-i}{k_B T} \frac{D_x K_{i,x} f_{d,i,x}^{(1)} + D_z K_{i,z} f_{d,i,z}^{(1)} + D_z K_{i,z} f_{r,i,z}^{(1)}}{D_x K_{i,x}^2 + D_z K_{i,z}^2 - i\delta}.$$

Finally, using the fact that the probe transmission spectrum is proportional to the imaginary component of $a_i^{(1)}$ and taking into account the dependence of $\mathbf{f}_{d,i}^{(1)}$ and $\mathbf{f}_{r,i}^{(1)}$ on the laser parameters and geometry, one finds that in the small angle limit

$$g_p \propto \left(D_x + D_z \frac{\theta^2}{4}\right) \frac{\gamma\delta}{\gamma^2 + \delta^2} + \left(\beta \frac{\Gamma}{\Delta} D_z\right) \frac{\gamma^2}{\gamma^2 + \delta^2}, \qquad (4)$$

where β is a numerical factor of the order of 1 dependent on the atomic transition. The first term in (4) can be traced back to the dipole force and contributes to the spectrum in the form of a conventional dispersive central resonance, whereas the second term arising from the radiation pressure effect along z has a Lorentzian contribution to the spectrum [15]. It is important to note that both structures have the same width $\gamma = k^2\theta^2(D_x + D_z\theta^2/4)$ corresponding to atomic spatial diffusion on the scale of the pump-probe interference periodicity (k: modulus of the probe and lattice beams wave vectors).

We now compare Eq. (4) against the results of our experiments. According to our simplified model, the shape of the central resonance has a dispersion to Lorentzian ratio $R = \Delta(D_x + D_z\theta^2/4)/\beta\Gamma D_z$ and thus evolves from a Lorentzian toward a dispersion as the frequency detuning of the lasers increases. We have performed a first series of experiments in the lattice geometry of Fig. 1 by varying the laser parameters I and Δ. In fact, it is generally more relevant physically to use two dimensionless quantities combining these parameters, namely, the optical potential depth (U_0) in units of the one-photon recoil energy $U_0/E_R \propto I/\Delta$ and the frequency detuning in units of the linewidth of the excited state Δ/Γ. We show in Fig. 2(a) the evolution of R versus Δ/Γ as U_0/E_R is kept constant ($U_0/E_R = 1200$), i.e., constant lattice topography. It clearly appears that the resonance tends to distort from a Lorentzian to a conventional dispersion shape as the frequency detuning increases. At the same time, γ decreases with the detuning before smoothly increasing in the limit of large detunings [Fig. 2(b)]. We also performed a series of experiments by varying independently the θ_x and θ_y angles of the tetrahedron in the range 20°–40°. We observed that to a good approximation, the width and the shape of the central resonance are only dependent on θ_x no matter the value of θ_y, as expected [16].

Finally, we have performed numerical simulations of the probe transmission spectrum using the semiclassical Monte Carlo simulation technique on the model case of a $J = 1/2 \to J' = 3/2$ transition [17,18]. The theoretical variations of R and γ shown in Figs. 2(c) and 2(d) appear to be in reasonable agreement with the experimental results [19], in spite of the difference between the model and the actual rubidium transition. In the course of these simulations, we checked that the Lorentzian shape resonance *disappears* when the term corresponding to the radiation pressure force along Oz is erased in the program. This is a supplementary proof of the physical origin of the resonance.

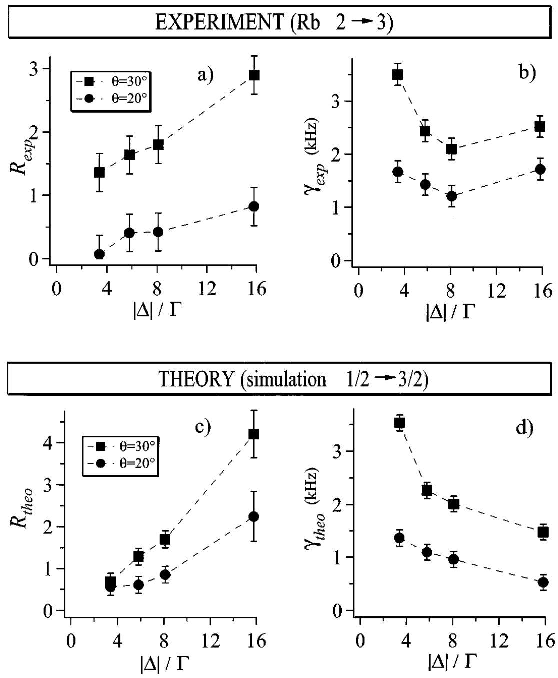

FIG. 2. Variation of the dispersion to Lorentzian ratio R and the width γ of the central resonance versus Δ/Γ. The data correspond to the same optical potential depth $U_0 = 1200E_R$. (a,b) Experimental points recorded for $\theta_x = \theta_y = 20°$ (full dots) and $\theta_x = \theta_y = 30°$ (full squares) in the case of the $2 \rightarrow 3$ transition of rubidium. (c,d) Results from a numerical simulation performed for the same angles in the model case of a $1/2 \rightarrow 3/2$ transition (γ_{theo} has been expressed in kHz using the recoil frequency of rubidium). The dashed lines are guides to the eye in all four figures.

In conclusion, we have observed a novel stimulated scattering process that shares several characteristics with the photorefractive effect. However, whereas the photorefractive effect relies on charged carriers and the electro-optic effect, radiation pressure-induced two-beam coupling involves the mechanical effect of photon absorption on neutral atoms. Still, the result presented in this Letter is a new example of the amazing analogies between condensed-matter systems and optical lattices.

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[1] See, for example, R.W. Boyd, *Nonlinear Optics* (Academic Press, New York, 1992), Sec. 6.4.
[2] N. Kukhtarev *et al.*, Ferroelectronics **22**, 949 (1979); **22**, 961 (1979).
[3] J. Feinberg *et al.*, J. Appl. Phys. **51**, 1297 (1980); **52**, 537 (1981).
[4] K.I. Petsas, A.B. Coates, and G. Grynberg, Phys. Rev. A **50**, 5173 (1994).
[5] T.W. Hodapp *et al.*, Appl. Phys. B **60**, 135 (1995); C. Jurczak *et al.*, Phys. Rev. Lett. **77**, 1727 (1996).
[6] P. Verkerk *et al.*, Europhys. Lett. **26**, 171 (1994).
[7] A. Kastberg *et al.*, Phys. Rev. Lett. **74**, 1574 (1995).
[8] J.-Y. Courtois and G. Grynberg, Phys. Rev. A **46**, 7060 (1992).
[9] D. Meacher *et al.*, Phys. Rev. Lett. **74**, 1958 (1995).
[10] G. Birkl *et al.*, Phys. Rev. Lett. **75**, 2823 (1995); M. Weidemuller *et al.*, Phys. Rev. Lett. **75**, 4583 (1995).
[11] J.-Y. Courtois *et al.*, Phys. Rev. Lett. **77**, 40 (1996).
[12] A detailed study of the central resonance in the case of a six-beam lattice and for small angles between probe and lattice beams was performed by A. Hemmerich *et al.*, Europhys. Lett. **22**, 89 (1994). The characteristics of their resonances are quite different from those reported here.
[13] Note that this model is somewhat oversimplified in that it does not take into account optical pumping processes and that it assumes isotropic thermal equilibrium in the lattice and a diagonal spatial diffusion tensor.
[14] The calculation makes use of a Fourier expansion. The coefficient $a_i^{(1)}$ is obtained through the component oscillating as $\exp[-i\delta t + i\mathbf{K}_i \cdot \mathbf{r}]$. In addition, the term $D_x K_{i,x} f_{r,i,x}^{(1)}$ is neglected compared to $D_z K_{i,z} f_{r,i,z}^{(1)}$ because in the present lattice geometry, $D_x \ll D_z$ as a result from the strong suppression of optical pumping in the x, y directions [Y. Castin *et al.*, Phys. Rev. A **50**, 5092 (1994)].
[15] It can be readily shown that the direction of the energy exchange between the frequency-degenerate pump and probe is only determined by the relative magnitude of components of their wave vectors along the symmetry axis, Oz.
[16] In the investigated range of laser parameters, the dependence of γ with θ is essentially compatible with a θ^2 law. This seems to indicate that the atomic density grating mainly relaxes through spatial diffusion along the x direction. In the framework of the simplified model it is possible to deduce D_x and D_z for each value of θ, U_0/E_R, Δ/Γ from the measurements of R and γ, provided, however, that the value of β is estimated, which is a difficult task in the case of the $2 \rightarrow 3$ atomic transition of our experiment.
[17] Y. Castin, J. Dalibard, and C. Cohen-Tannoudji, in *Light-Induced Kinetic Effects on Atoms, Ions and Molecules,* edited by L. Moi *et al.* (ETS Editrice, Pisa, 1991).
[18] J.-Y. Courtois, in *Coherent and Collective Interactions of Particles and Radiation Beams,* Proceedings of the International School of Physics "Enrico Fermi," Course CXXXI, edited by A. Aspect, W. Barletta, and R. Bonifacio (IOS Press, Amsterdam, 1996), p. 341.
[19] We attribute the discrepancy between theory and experiment in the large detuning behavior of γ to the difference in the atomic transitions. In particular, the multipotential nature of laser cooling typical of transitions with large angular momenta like in rubidium are not accounted for in our $1/2 \rightarrow 3/2$ model. The increasing influence of the $F = 2 \rightarrow F' = 2$ atomic transition of rubidium as the laser detuning increases may also affect the results.