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## Influence of Nearly Resonant Light on the Scattering Length in Low-Temperature Atomic Gases

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We develop the idea of manipulating optically the scattering length  $a$  in low-temperature atomic gases. If the incident light is close to resonance with one of the bound  $p$  levels of electronically excited molecules, virtual radiative transitions of a pair of interacting atoms to this level can significantly change the value and even reverse the sign of  $a$ . The decay of the gas due to photon recoil and due to photoassociation can be minimized by selecting the frequency detuning and the Rabi frequency. Our calculations show the feasibility of optical manipulations of trapped Bose condensates through a light-induced change in the mean field interaction between atoms, which is illustrated for  $^7\text{Li}$ . [S0031-9007(96)01316-6]

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The recent successful experiments on Bose-Einstein condensation (BEC) in magnetically trapped gases of Rb [1], Li [2], and Na [3] have generated a lot of interest in macroscopic quantum behavior of atomic gases at ultralow temperatures. These experiments were enabled by efficient evaporative [4] and optical cooling [5,6] methods combined to reach the necessary temperatures ( $T \lesssim 1 \mu\text{K}$ ) and densities  $10^{12} \lesssim n \lesssim 10^{14} \text{ cm}^{-3}$ .

A principal question for BEC in atomic gases concerns the sign of the scattering length  $a$  for the pair elastic interaction. For  $a > 0$ , elastic interaction between atoms is repulsive, and the Bose condensate is stable with respect to this interaction. If  $a < 0$ , elastic interaction is attractive, and this is the origin of a collapse of the condensate in a homogeneous gas [7]. For trapped gases with  $a < 0$  the situation is likely to be the same, provided the interaction between particles exceeds the level spacing in the trapping field [8,9]. If this interaction is much smaller than the level spacing, there is a gap for one-particle excitations, and it is possible to form a metastable Bose-condensed state [9]. Among the alkalis, there are atomic gases with positive as well as with negative  $a$  [10]. Also, the magnetic field dependence of  $a$  was predicted [11].

In this Letter we develop the idea of manipulating the value and the sign of the scattering length by using nearly resonant light. Since changing  $a$  directly affects the mean field interaction between the atoms, this offers a possibility to investigate macroscopic quantum phenomena associated with BEC by observing the evolution of a Bose-condensed gas in response to light. The physical picture of the influence of the light field on the elastic interaction between atoms is the following: A pair of atoms absorbs a photon and undergoes a virtual transition to an electronically excited quasimolecular state. Then it reemits the photon and returns to the initial electronic state at the same kinetic energy. As the interaction between

atoms in the excited state is much stronger than in the ground state, at moderate light intensities the scattering amplitude can already be significantly changed. Aside from the collective effects, as discussed here, optical control of the rates of intrinsic inelastic collisions [6,12] and elastic collisional rates [13] has attracted a lot of interest.

The presence of the light field also induces inelastic processes, such as photon recoil and light absorption in pair collisions (with regard to cold collisions, see Refs. [14] and [15] for review). Photon recoil is the result of the scattering of light by single atoms. At subrecoil temperatures, typical for achieving BEC, recoiling atoms are lost as they overcome the confining barrier and escape from the trap. The probability of light scattering by single atoms is proportional to  $(\Omega/\delta)^2$ , where  $\Omega$  is the Rabi frequency and  $\delta$  is the frequency detuning of the light with respect to a single atom at rest. To suppress the recoil losses the ratio  $\Omega/\delta$  needs to be sufficiently small. Then, for positive  $\delta$ , where the light is at resonance with continuum states of excited quasimolecules, the change of  $a$  will also be proportional to  $(\Omega/\delta)^2$  and thus very small. To have small recoil losses in combination with a significant change of  $a$ , the detuning  $\delta$  should be large and negative and not too far from a vibrational resonance with one of the bound  $p$  states of the electronically excited molecule. However, the vicinity of the resonance will also lead to photoassociation in pair collisions, followed by spontaneous emission and loss from the trap. Hence, the frequency detuning  $\delta_\nu$  with respect to the  $\nu$ th (nearest) vibrational resonance should greatly exceed the linewidth of the resonance. We established that it is possible to change the scattering length substantially and even switch its sign without excessive recoil or photoassociation losses. This will be illustrated for  $^7\text{Li}$ .

We consider low gas densities satisfying the condition

$$n\lambda^3 \ll 1, \quad (1)$$

where  $2\pi\lambda$  is the optical wavelength. Then collective optical effects [16] are absent, and at sufficiently low temperatures the line broadening of optical transitions is determined by the natural linewidth  $\Gamma = 4d^2/3\hbar\lambda^3$ , where  $d$  is the transition dipole moment. We analyze the influence of incident light with large ( $|\delta| \gg \Gamma$ ) and negative frequency detuning on the interaction in a pair of atoms, with vanishing wave vector of relative motion,  $\mathbf{k} \rightarrow 0$ . The light frequency is assumed to be nearly resonant with a highly excited vibrational  $p$  level (with vibrational quantum number  $\nu$ , rotational quantum number  $J = 1$ , and binding energy  $\varepsilon_\nu$ ) in the interaction potential  $V(r)$  of the attractive excited electronic state of the quasimolecule, i.e., the frequency detuning with respect to this level,  $\delta_\nu = \delta - \varepsilon_\nu$ , is much smaller than the local vibrational level spacing  $\Delta\varepsilon_\nu = \varepsilon_\nu - \varepsilon_{\nu+1}$  (hereafter, all frequencies are given in energy units). Then radiative transitions of the pair from the ground electronic state to the excited level  $\nu$  are most important. These transitions occur predominantly at interparticle distances  $r$  in the vicinity of the outer turning point  $r_t$  for the relative motion of atoms in the bound state  $\nu$ , i.e.,  $V(r_t) = -\varepsilon_\nu$ . Unless  $\varepsilon_\nu$  is very large,  $r_t$  is determined by the long-range part of  $V(r)$ , represented by the resonance dipole term. If  $\varepsilon_\nu$  and  $|\delta|$  are still much larger than the Zeeman and fine structure splitting, then, at interparticle distances relevant for radiative transitions, the polarization vector of the attractive excited quasimolecular state,  $\mathbf{e}_\lambda$ , is parallel to the internuclear axis, and  $V(r) = -2d^2/r^3$ . Hence, as  $\varepsilon_\nu \sim |\delta| \gg \Gamma$ , we have  $r_t \ll \lambda$ .

For sufficiently large  $\varepsilon_\nu$  and  $\delta$ , spontaneous emission of excited molecules predominantly produces nontrapped atoms with kinetic energies of order  $\varepsilon_\nu$ . These atoms practically do not interact with the driving light and escape from the trap. Therefore, the problem of finding the scattering length in the presence of light is equivalent to a scattering problem which can be described in terms of wave functions of the ground and excited electronic quasimolecular states. These states are coupled by light, and spontaneous emission from the excited state can be taken into account by adding the "absorptive part"  $-i\Gamma$  (the spontaneous emission rate for molecules is twice as large as that for single atoms) to the interaction potential  $V(r)$ .

In the Born-Oppenheimer approximation the total wave function of the quasimolecule in the presence of light can be written as  $\phi(\mathbf{r})|g\rangle + \psi(\mathbf{r})|e\rangle$ , where  $|g\rangle$  and  $|e\rangle$  are the electron wave functions of the ground and excited electronic states. The wave functions of the relative motion of atoms in these states,  $\phi(\mathbf{r})$  and  $\psi(\mathbf{r})$ , can be found from the system of coupled Schrödinger equations:

$$-\frac{\hbar^2}{m} \Delta_{\mathbf{r}} \phi(\mathbf{r}) + U(r)\phi(\mathbf{r}) + \Omega \xi(\mathbf{r})\psi(\mathbf{r}) = 0, \quad (2)$$

$$-\frac{\hbar^2}{m} \Delta_{\mathbf{r}} \psi(\mathbf{r}) + [V(r) - i\Gamma - \delta]\psi(\mathbf{r}) + \Omega \xi(\mathbf{r})\phi(\mathbf{r}) = 0, \quad (3)$$

where  $\xi(\mathbf{r}) = \mathbf{e}_\alpha \mathbf{e}_\lambda(\mathbf{r})$ ,  $U(r)$  is the interaction potential in the ground electronic state, and  $\mathbf{e}_\alpha$  the polarization vector of light. The Rabi frequency is defined as  $\Omega = dE/\sqrt{2}$ , where  $E$  is the amplitude of the electric field of light. In Eqs. (2) and (3) we neglect the light shifts at infinite separation between atoms and omit the recoil. These equations lead to the following integral equation for  $\phi(\mathbf{r})$ :

$$\phi(\mathbf{r}) = \phi_0(\mathbf{r}) + \Omega^2 \int d\mathbf{r}'' d\mathbf{r}' G(\mathbf{r}'', \mathbf{r}) \xi(\mathbf{r}'') \tilde{G}(\mathbf{r}'', \mathbf{r}') \times \xi(\mathbf{r}') \phi(\mathbf{r}'). \quad (4)$$

Here  $G(\mathbf{r}, \mathbf{r}')$  and  $\tilde{G}(\mathbf{r}, \mathbf{r}')$  are the Green functions of Eqs. (2) and (3) with  $\Omega = 0$ . The wave function  $\phi_0$  describes the relative motion of atoms with zero energy for the potential  $U(r)$  in the absence of light. This function is a solution of Eq. (2) with  $\Omega = 0$ . The Green function  $G(\mathbf{r}, \mathbf{r}')$  has the form

$$G(\mathbf{r}, \mathbf{r}') = \frac{m}{4\pi\hbar^2} \times \begin{cases} \phi_0(r)\tilde{\phi}_0(r'), & r < r' \\ \phi_0(r')\tilde{\phi}_0(r), & r > r' \end{cases}, \quad (5)$$

where  $\tilde{\phi}_0(r)$  is a solution of the same Schrödinger equation as that for  $\phi_0(r)$ , but contains only an outgoing spherical wave at large  $r$ :  $\tilde{\phi}_0(r) \rightarrow 1/r$  for  $r \rightarrow \infty$ . As the frequency detuning of light was chosen such that  $|\delta_\nu| \ll \Delta\varepsilon_\nu$ , the bound state  $\nu$  should give the dominant contribution to  $\tilde{G}(\mathbf{r}, \mathbf{r}')$ , and we may use

$$\tilde{G}(\mathbf{r}, \mathbf{r}') = -\psi_\nu(\mathbf{r})\psi_\nu^*(\mathbf{r}')/(\delta_\nu + i\Gamma), \quad (6)$$

where  $\psi_\nu(\mathbf{r})$  is the wave function of this state in the absence of light. Accordingly, the dependence of the rhs of Eq. (4) on  $\phi(\mathbf{r})$  will be contained only in the integral  $I = \int d\mathbf{r}' \phi(\mathbf{r}') \xi(\mathbf{r}') \psi_\nu^*(\mathbf{r}')$ . Multiplying both sides of Eq. (4) by  $\xi(\mathbf{r}) \psi_\nu^*(\mathbf{r})$  and integrating over  $d\mathbf{r}$ , we express  $I$  through the overlap integral  $I_0 = \int d\mathbf{r}' \phi_0(\mathbf{r}') \xi(\mathbf{r}') \psi_\nu^*(\mathbf{r}')$ . Then the exact solution of Eq. (4) is straightforward:

$$\phi(\mathbf{r}) = \phi_0(\mathbf{r}) - \frac{\Omega^2 I_0 \int d\mathbf{r}' \psi_\nu(\mathbf{r}') \xi(\mathbf{r}') G(\mathbf{r}, \mathbf{r}')}{\delta_\nu + (\Omega^2/\Delta\varepsilon_\nu)\beta + i\Gamma}. \quad (7)$$

The quantity  $(\Omega^2/\Delta\varepsilon_\nu)\beta$  describes the light-induced shift of the  $\nu$ th vibrational resonance, and the numerical factor  $\beta = \Delta\varepsilon_\nu \int d\mathbf{r} d\mathbf{r}' G(\mathbf{r}, \mathbf{r}') \xi(\mathbf{r}) \psi_\nu^*(\mathbf{r}') \xi(\mathbf{r}') \psi_\nu(\mathbf{r}')$ . As in the limit of zero energies only the  $s$ -wave contribution to  $\phi(\mathbf{r})$  and  $\phi_0(\mathbf{r})$  is important, the scattering length in the presence of light can be found from the asymptotic form of  $\phi(\mathbf{r})$  at large distances:  $\phi(\mathbf{r}) \rightarrow 1 - a/r$  for  $r \rightarrow \infty$ . At large  $r$ , the Green function  $G(\mathbf{r}, \mathbf{r}') = m\phi_0(\mathbf{r}')/4\pi\hbar^2 r$ , and Eq. (7) yields

$$a = \bar{a} + \frac{(\Omega^2/\Delta\varepsilon_\nu)\tilde{\beta}}{\delta_\nu + (\Omega^2/\Delta\varepsilon_\nu)\beta + i\Gamma} r_t, \quad (8)$$

with  $\bar{a}$  the scattering length in the absence of light, and the numerical factor  $\tilde{\beta} = (m\Delta\varepsilon_\nu/4\pi\hbar^2 r_t)|I_0|^2$ . It should be emphasized that Eq. (8) is valid for any ratio between  $|\delta_\nu|$  and  $(\Omega^2/\Delta\varepsilon_\nu)\beta$ .

The main contribution to the integrals in the equations for  $\beta$ ,  $I_0$ , and  $\tilde{\beta}$  originates from distances  $r$  in the vicinity of the turning point  $r_t$ . Unless  $\varepsilon_\nu$  and  $|\delta|$  are huge and  $r_t$  is small,  $\phi_0$  and  $\tilde{\phi}_0$  are smooth functions of  $r$  at distances

$r \sim r_t$ , and one may put  $\phi_0(r) = \phi_0(r_t)$ ,  $\tilde{\phi}_0(r) = \tilde{\phi}_0(r_t)$  in the integrands. The excited-state wave function  $\psi_\nu(\mathbf{r})$  can be found within the WKB approach, with a standard modification using a linear approximation for the potential  $V(r) = -2d^2/r^3$  at  $r$  close to  $r_t$ . This gives  $\psi_\nu(\mathbf{r})$  at  $r$  near  $r_t$  in the form of an Airy function, and we obtain

$$\tilde{\beta} = 0.8\pi^2\phi_0^2(r_t), \quad \beta = 0.8\pi^2f_0(r_t)\phi_0(r_t). \quad (9)$$

The function  $f_0(r) = r\tilde{\phi}_0(r)$  is tending to 1 for  $r \rightarrow \infty$ . Direct calculations for  ${}^7\text{Li}$  show that Eq. (9) is accurate within a few percent for transitions to vibrational states with  $\nu \geq 60$ . For the level spacing, the WKB approximation gives

$$\Delta\varepsilon_\nu = 1.9\pi\varepsilon_\nu(r_t/r_0)^{1/2} \ll \varepsilon_\nu. \quad (10)$$

The characteristic distance  $r_0 = md^2/\hbar^2$ . For alkali atoms,  $r_0$  greatly exceeds the optical wavelength ( $r_0 \geq 10^5 \text{ \AA}$ ) and, hence,  $r_0 \gg \lambda \gg r_t$ .

The presence of other bound  $p$  levels and continuum states of the excited quasimolecule changes Eq. (6) for the Green function  $\tilde{G}$ . Our analysis, relying on the exact expression for  $\tilde{G}$ , shows that, in order to omit the contribution of virtual transitions to these states and, hence, retain the validity of Eq. (8), it is sufficient to have  $|\delta_\nu|$  and  $\Omega$  much smaller than the level spacing  $\Delta\varepsilon_\nu$ . The condition  $\Omega \ll \Delta\varepsilon_\nu$  leads to important physical consequences. The radiative transitions occur in a narrow range of distances near  $r_t$ , characterized by the width  $\Delta r \sim r_t(r_t/r_0)^{1/3}$ . Accordingly, the characteristic separation between the unperturbed potential curves  $U(r)$  and  $V(r)$  in this region is  $\varepsilon_\nu(\Delta r/r_t) \sim \Delta\varepsilon_\nu(r_0/r_t)^{1/6} \gg \Omega$ . This inequality ensures the absence of effects analogous to power broadening in the single atom case.

The light changes the real part of the scattering length and introduces an imaginary part. The frequency dependence of  $\text{Re}a$  and  $\text{Im}a$  has a resonance structure:

$$\begin{aligned} \text{Re}a &= \bar{a} + \frac{\Omega^2\tilde{\beta}\zeta_\nu}{\Delta\varepsilon_\nu(\zeta_\nu^2 + \Gamma^2)}r_t, \\ \text{Im}a &= -\frac{\Omega^2\tilde{\beta}\Gamma}{\Delta\varepsilon_\nu(\zeta_\nu^2 + \Gamma^2)}r_t, \end{aligned} \quad (11)$$

where  $\zeta_\nu = \delta_\nu + (\Omega^2/\Delta\varepsilon_\nu)\beta$ . The real part determines the mean field interaction between atoms. The light-induced change of this interaction is given by

$$n(\tilde{U} - \bar{U}) \equiv \hbar\tau_a^{-1} = 4\pi\hbar^2(\text{Re}a - \bar{a})n/m. \quad (12)$$

The imaginary part of  $a$  is a consequence of spontaneous emission from the excited molecular state  $\nu$  formed in the process of light absorption in pair collisions (photoassociation). The inverse decay time due to this process is

$$\tau_{pa}^{-1} = 8\pi\hbar|\text{Im}a|n/m. \quad (13)$$

Exactly at resonance ( $\zeta_\nu = 0$ ) the mean field interaction is the same as in the absence of light, and the photoassociation rate is the largest.

For small Rabi frequency, Eq. (11) goes over into the result of perturbation theory, and both  $\tau_a^{-1}$  and  $\tau_{pa}^{-1}$  are

proportional to  $\Omega^2$ . The former can be treated as a "light shift" of the mean field interaction, and the latter will be nothing more than the ordinary photoassociation rate at a low light power. For  $(\Omega^2/\Delta\varepsilon_\nu)\beta \gg |\delta_\nu|$  the driving light shifts the interacting pair of atoms out of resonance. As the corresponding shift is proportional to  $\Omega^2$ , the light-induced change of the mean field interaction becomes independent of  $\Omega$ . It will be determined by Eq. (12) with  $\text{Re}a - \bar{a} = (\tilde{\beta}/\beta)r_t$ . On the other hand, the photoassociation rate ( $\text{Im}a$ ) decreases as  $1/\Omega^2$ .

The amplitude of binary interaction, affected by light, undergoes damped oscillations and, in any case, reaches its stationary value (8) on a time scale of order  $\Gamma^{-1}$ . This is much shorter than the characteristic response time of a dilute trapped gas, which cannot be faster than  $\tau_a$ . To have an appreciable modification of the mean field interaction without excessive photoassociation,  $\tau_a$  should be short compared to  $\tau_{pa}$ , i.e., the condition

$$|\text{Re}a| \gg |\text{Im}a| \quad (14)$$

should hold. As follows from Eq. (11), this is the case for  $|\zeta_\nu| \gg \Gamma$ . The change of the scattering length  $\text{Re}a - \bar{a} \approx (\Omega^2\tilde{\beta}/\Delta\varepsilon_\nu\zeta_\nu)r_t$  can exceed  $r_t$ , and the scattering length can be changed in both directions simply by changing the sign of  $\zeta_\nu$ .

In addition, the time  $\tau_a$  should be much smaller than the decay time  $\tau_r$  due to the photon recoil of single atoms, caused by light scattering. Since  $\tau_r^{-1} = (\Omega/\delta)^2\Gamma/2$ , this is the case for ( $|\delta| \approx \varepsilon_\nu$ )

$$n\lambda^3 \gg |\zeta_\nu|/4\Delta\varepsilon_\nu, \quad (15)$$

as follows from Eqs. (9)–(11), assuming  $|\zeta_\nu| \gg \Gamma$  to simultaneously satisfy condition (14). As  $|\zeta_\nu| \ll \Delta\varepsilon_\nu$ , the inequality (15) is not in contradiction with our starting assumption (1) and can be fulfilled in alkali atom gases at densities  $n \sim 10^{13} - 10^{14} \text{ cm}^{-3}$  by an appropriate choice of  $\Omega$ , the level  $\nu$ , and  $\delta_\nu$ .

All the above results remain valid for finite momenta of colliding atoms,  $k \ll \min\{r_t^{-1}, |\bar{a}|^{-1}, |a|^{-1}\}$ .

We performed calculations for  ${}^7\text{Li}$  by using  $\Gamma = 3.7 \times 10^7 \text{ s}^{-1}$  and spectroscopic information on the location of bound  $p$  levels in the excited electronic state  ${}^3\Sigma_g^+$  [17]. The functions  $\phi_0(r)$  and  $f(r)$  were calculated by using the potential of interaction in the ground state  ${}^3\Sigma_u^+$  from [18], the scattering length in the absence of light being  $\bar{a} \approx -14 \text{ \AA}$ . Equation (11) was used to calculate the scattering length  $a$  under the influence of light nearly resonant for vibrational  $p$  levels of the  ${}^3\Sigma_g^+$  state, with quantum numbers ranging from  $\nu = 77$  ( $\varepsilon_\nu = 2.8 \text{ K}$ ) to  $\nu = 66$  ( $\varepsilon_\nu = 28.7 \text{ K}$ ). We find that, for  $\Omega$  in the range 5–40 mK (light power from 10 to 1000 W/cm<sup>2</sup>), it is possible to significantly change the scattering length and even make it positive while maintaining  $|\text{Im}a| \ll |\text{Re}a|$  (see Fig. 1). The recoil loss time  $\tau_r$  varies from 100 to 1 ms.

Our results show the feasibility to optically manipulate the mean field interaction between atoms and open

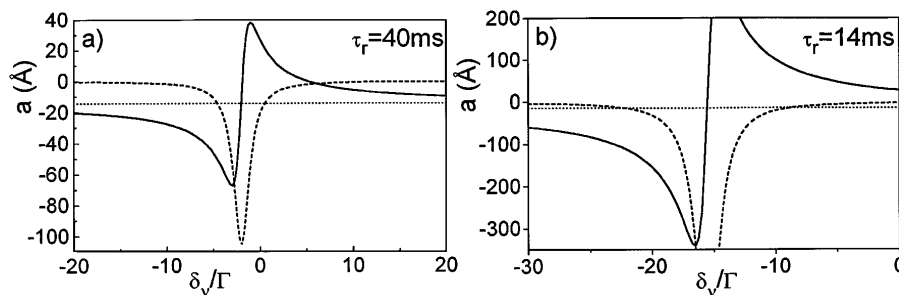


FIG. 1. The scattering length for  ${}^7\text{Li}$  as a function of the frequency detuning of light,  $\delta_\nu/\Gamma$ , from the excited bound  $p$  level  $\nu$ : (a)  $\Omega = 10$  mK,  $\varepsilon_\nu = 9.1$  K; (b)  $\Omega = 40$  mK,  $\varepsilon_\nu = 20.1$  K. The solid curve represents the real part of  $a$ , and the dashed curve the imaginary part. The dotted line corresponds to the scattering length in the absence of light.

prospects for new optical experiments in trapped gases. For example, once a gas is in a Bose-condensed state, instantaneous switching of the sign of  $a$  changes the sign of the nonlinear interaction term in the Ginzburg-Gross-Pitaevskii equation for the condensate wave function and causes the trapped condensate to evolve in a completely different way than a condensate set into motion by changing the trap frequency. The evolution will involve two time scales:  $\tau_a$  and the inverse trap frequency  $\omega_t^{-1}$ , and continue after the light is switched off. Because of the light-induced decay processes, the light should be switched on only for a time much shorter than  $\tau_r$ . Hence, besides the above discussed condition  $\tau_a \ll \tau_r$ , experiments should be arranged such that  $\omega_t \tau_r \gg 1$ . This is feasible with the above values for  $\tau_r$ . As in most cases,  $\tau_r$  will be much smaller than the characteristic time for elastic collisions, the evolving condensate will not be in equilibrium with above-condensate particles.

In trapped gases with negative scattering length, one may expect a stabilization of the condensate by switching  $a$  to positive values. Of particular interest is the case where the sign of  $a$  is switched from positive to negative. In a quasihomogeneous Bose-condensed gas ( $n\bar{U} \gg \hbar\omega_t$ ) this should induce a ‘‘collapse’’ of the condensate, caused by elastic interatomic interaction. The investigation of this phenomenon is of fundamental interest.

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