Light-induced ground-state coherence, constructive interference, and two-photon laser cooling mechanism in multilevel atoms

Soo Chang
Department of Physics, Hannam University, 133 Ojungdong, Taejon, Korea

Taeg Yong Kwon and Ho Seong Lee
Division of Electromagnetic Metrology, Korea Research Institute of Standards and Science, P.O. Box 102, Yusong, Taejon, Korea

Vladimir Minogin
Institute of Spectroscopy Russian Academy of Sciences, 142092 Troitsk, Moscow Region, Russia and Korea Research Institute of Standards and Science, P.O. Box 102, Yusong, Taejon 305-600, Korea

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It is shown that dipole interaction of multilevel atoms having three or more ground-state sublevels with counterpropagating circular polarized laser waves is strongly influenced by ground-state coherence produced by two-photon processes. In the simplest case of a (3+5)-level atom the ground-state coherence is shown to be responsible for constructive atomic interference that enhances upper atomic populations at small velocities. Ground-state coherence is also responsible for the coherent redistribution of the ground-state atomic populations at small velocities. It is found that a two-photon coherent redistribution of atomic populations enhances considerably the radiation force at small velocities and accordingly the friction produced by the radiation force. [S1050-2947(99)09109-X]

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Atomic-state coherence plays an important role in the processes of the dipole interaction of atoms with laser fields. It is well known that in the case of the three-level Λ-interaction scheme, the ground-state coherence is responsible for the coherent population trapping of atoms in the ground-state sublevels and accordingly for the destructive interference of the two dipole excitation channels that results in no atomic excitation to the upper state [1–3]. While the Λ-interaction scheme is a very specific one, there is a broad class of dipole interaction schemes that can be considered as $V\cdots V$-interaction schemes. Such interaction schemes describe the optical excitation of atoms by circularly polarized laser waves on the transitions between hyperfine structure magnetic sublevels $|F,m_F\rangle$ in cases when the upper-state degeneracy $2F_e+1$ exceeds the ground-state degeneracy $2F_g+1$. These schemes are nowadays of broad use in experiments on laser cooling of alkali-metal atoms in atomic vapors or in magnetic and magneto-optical traps [4–7].

In commonly used one-, two-, or three-dimensional $V\cdots V$-interaction schemes atoms are irradiated by pairs of counterpropagating laser waves having the same circular polarization, left- or right-hand polarization. When polarizations of the two counterpropagating waves are considered with respect to the quantization axis that coincides with the common propagation axes, the laser field configuration is usually denoted as a σ^+-σ^- configuration.

Since any $V\cdots V$ scheme includes at least one Λ scheme, the interaction of atoms with σ^+-σ^- laser configuration generally speaking depends strongly on the value and sign of the ground-state coherence produced by two-photon processes. The behavior of the ground-state coherence and its velocity dependence in the case of a $V\cdots V$ scheme differs considerably, however, from that for the Λ scheme due to the additional strong left- and right-hand excitation channels.

It is the purpose of this paper to show that contrary to the Λ-interaction schemes, the ground-state coherence produced by two-photon processes in the $V\cdots V$-interaction schemes can have an opposite sign at small velocities and can thus be responsible not for destructive but *constructive interference* of the dipole excitation channels. Besides the constructive interference the ground-state coherence produces a *coherent redistribution* of the ground-state populations at small velocities. In the simplest case of a (3+5)-level atom, considered below, constructive interference produces an increased atomic population in the upper central state and due to spontaneous decays also produces an increased population in the central ground state. Populations of the left- and right-hand ground-state sublevels undergo dispersive-type coherent redistribution in the zero-velocity region.

An important consequence of the coherent redistribution of the ground-state atomic population is the *enhancement of the radiation force at small velocities* and the friction produced by the radiation force at a negative detuning. The two-photon processes that produce ground-state coherence in $V\cdots V$-interaction schemes can thus be attributed to a physical mechanism responsible for the deep cooling of alkali-metal atoms in counterpropagating σ^+-σ^- laser waves.

The simplest example of a $V\cdots V$-interaction scheme that includes three ground-state atomic sublevels ($F_g=1$) and five upper-state sublevels ($F_e=2$) is shown in Fig. 1. In this VVV-interaction scheme, a (3+5)-level atom is assumed to interact with two counterpropagating left-circularly polarized laser waves,

$$\mathbf{E}=\mathbf{E}_1+\mathbf{E}_2,$$

$$\mathbf{E}_1=\frac{E_0}{2}[\mathbf{e}_+e^{i(kz-\omega t)}-\mathbf{e}_-e^{-i(kz-\omega t)}],$$

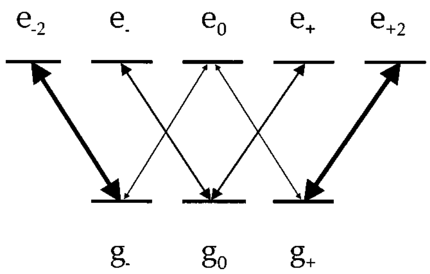

FIG. 1. Scheme of a (3+5)-level atom interacting with counter-propagating $\sigma^{\pm}$ polarized laser waves. Note that the interaction scheme includes three V schemes that start from the ground-state sublevels g_-, g_0, g_+. The widths of the lines showing dipole transitions are chosen to be proportional to the squares of the dipole matrix elements.

$$\mathbf{E}_2=\frac{E_0}{2}[-\mathbf{e}_+e^{i(kz+\omega t)}+\mathbf{e}_-e^{-i(kz+\omega t)}], \tag{1}$$

where $\mathbf{e}_\pm=\mp(1/\sqrt{2})(\mathbf{e}_x\pm i\mathbf{e}_y)$ are the spherical unit vectors, $k=\omega/c$ is the magnitude of a wave vector, and ω is the frequency of the laser waves. With respect to the quantization axis Oz the first wave is a σ^+-polarized wave and the second one is a σ^--polarized wave.

In the above scheme, the strength of the dipole interaction can be described by the value of the Rabi frequency at most strong left- or right-hand transitions, $\Omega=\|d\|E_0/2\sqrt{5}\hbar$, where $\|d\|$ is the reduced dipole matrix element, the detuning of the laser field frequency with respect to the atomic transition frequency, $\delta=\omega-\omega_0$, and the Doppler shift kv. For the physically most important case of low one-photon saturation, the interaction of an atom with the field is mainly determined by one-photon and two-photon processes and depends mainly on small one-photon effective saturation parameters $s_\pm=\Omega^2/[\gamma^2+(\delta\pm kv)^2]\ll 1$, where γ is a half of the total spontaneous decay rate at most left or most right atomic transitions, $W_{sp}=2\gamma=4\|d\|^2\omega^3/3\hbar c^3$. These processes produce optical coherence $\rho_{g_m e_n}$ between ground- and excited-state sublevels and ground-state coherence $\rho_{g_-g_+}$ that modifies atomic populations.

For slowly moving atoms, $kv\ll\gamma$, weakly saturated by the laser field, the equations for steady-state atomic density matrix elements $\rho_{\alpha\beta}$ can be simplified to a set of equations for ground-state populations $N_\alpha=\rho_{g_\alpha g_\alpha}$, $\alpha=-1,0,1$, and ground-state coherence $\nu=\rho_{g_-g_+}\exp(2ikz)$:

$$N_-+N_+=\frac{9}{2}N_0-(\nu+\nu^*),\quad N_--N_+=i\frac{\delta}{\gamma}(\nu-\nu^*),$$
$$(\mu-ikv)\nu=\frac{\Omega^2}{12}\left[\gamma(N_-+N_+)+i\delta(N_--N_+)+\frac{3\gamma}{\gamma^2+\delta^2}N_0\right],$$
$$N_-+N_0+N_+=1, \tag{2}$$

where quantity $\mu=(5/6)s\gamma$ determines the decay rate for the ground-state coherence induced by dipole transitions and $s=\Omega^2/(\gamma^2+\delta^2)$ is the effective saturation parameter at zero velocity, $s\ll 1$. The above equations directly follow from the total set of the density matrix equations if one excludes from the equations the off-diagonal density matrix elements and upper-state populations $n_\alpha=\rho_{e_\alpha e_\alpha}$: $n_{-2}=s\ N_-$, $n_-=n_+=sN_0/2$, $n_0=3sN_0/4$, $n_2=sN_+$. The ground-state coherence ν cannot be neglected in Eqs. (2), because it has the same order of magnitude as the ground-state populations. The last equation in set (2) is a normalization condition in the low-saturation limit.

Note that the neglect of the upper-state populations in Eqs. (2), including the normalization condition, introduces an error of the order of s into the ground-state populations (which have the order of magnitude 1) and an error of the order of s^2 into the upper-state populations (which have the order of magnitude s). The last error thus cannot change the order of magnitude of the ground-state and upper-state populations.

The solution to the above equations gives ground-state populations and ground-state coherence in lowest orders in a small effective saturation parameter $s\ll 1$ and a small velocity parameter $\eta=kv/\gamma$:

$$N_-=\frac{1}{2\Delta}\left[9+\frac{3}{5}\frac{\mu^2}{\mu^2+k^2v^2}\left(\frac{13}{5}\frac{\delta^2}{\gamma^2}-2\right)-\frac{3\mu kv}{\mu^2+k^2v^2}\frac{\delta}{\gamma}\right],$$
$$N_0=\frac{2}{\Delta}\left[1+\frac{1}{5}\frac{\mu^2}{\mu^2+k^2v^2}\left(\frac{6}{5}\frac{\delta^2}{\gamma^2}+1\right)\right],$$
$$N_+=\frac{1}{2\Delta}\left[9+\frac{3}{5}\frac{\mu^2}{\mu^2+k^2v^2}\left(\frac{13}{5}\frac{\delta^2}{\gamma^2}-2\right)+\frac{3\mu kv}{\mu^2+k^2v^2}\frac{\delta}{\gamma}\right],$$
$$\nu=\frac{3}{2\Delta}\frac{\mu}{\mu^2+k^2v^2}\left[\left(1+\frac{1}{5}\frac{\delta^2}{\gamma^2}\right)\mu+ikv\right], \tag{3}$$

where the common denominator describes saturation due to two-photon transitions,

$$\Delta=11+\frac{1}{5}\frac{\mu^2}{\mu^2+k^2v^2}\left(\frac{51}{5}\frac{\delta^2}{\gamma^2}-4\right). \tag{4}$$

The above solution shows that even at a low one-photon saturation the ground-state populations include narrow velocity structures due to the two-photon processes. In the low-velocity region, the contributions of these narrow structures are proportional to the Lorentzian forms that include the second power of the laser intensity both in the numerator and denominator. Accordingly, at low velocities these contributions have exactly the same order of magnitude as the contributions due to the one-photon processes. The narrow structures are located at zero velocity and they influence strongly both atomic populations and optical coherences at low velocities. The ground-state coherence ν created by two-photon processes is responsible for a narrow peak in population n_0 of the central upper state (Fig. 2). This structure demonstrates constructive atomic interference at small velocities. Spontaneous decay of the peak in the population n_0 can be shown to be responsible in turn for a narrow peak in the

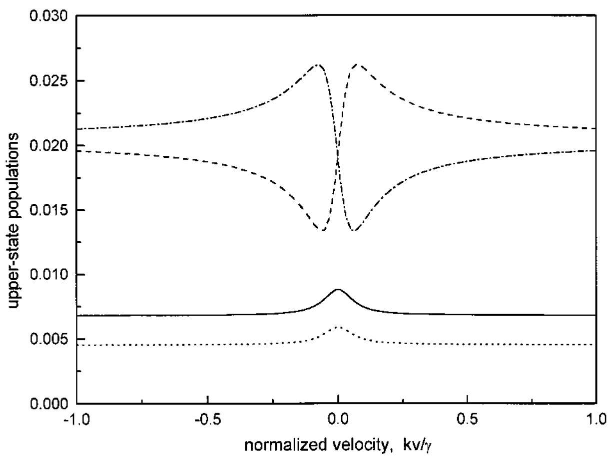

FIG. 2. Upper-state atomic populations n_{-2} (dashed line), $n_- = n_+$ (dotted line), n_0 (solid line), and n_{+2} (dash-dotted line) as functions of velocity for saturation parameter $G=1$ and detuning $\delta=-3\gamma$.

population of the central ground state N_0 (Fig. 3). If one excludes from the density matrix equations the spontaneous decay from the central upper state to the central ground state, the peak in the central ground state disappears. The central ground-state population is coupled to the upper-state populations n_- and n_+ mainly by one-photon transitions which are responsible for the narrow peaks in populations n_- and n_+. Other ground- and upper-state populations include dispersive-type structures caused by the coherent redistribution of the atomic population.

The qualitative behavior of the ground-state coherence and populations at small velocities can be described by simple physical arguments. The velocity position of the narrow two-photon structures can be directly estimated from the energy conservation law. In the atom rest frame, the absorption of a photon from one traveling wave and the emission of a photon to the other traveling wave results in a transition

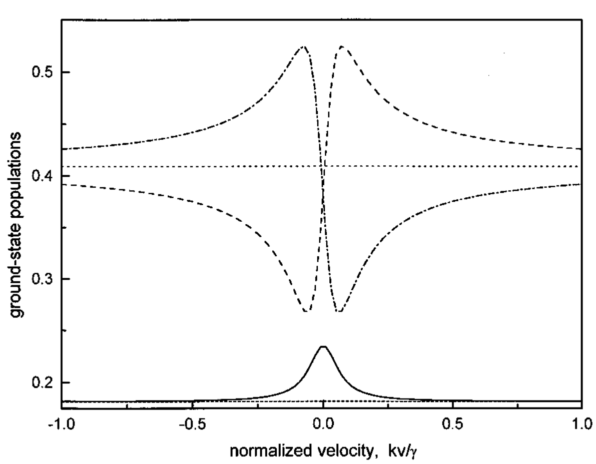

FIG. 3. Ground-state populations N_- (dashed line), N_0 (solid line), and N_+ (dash-dotted line) as functions of velocity for the same parameters as in Fig. 2. Dotted lines show values of ground-state populations in a limit of zero saturation: $N_0=4/22$, $N_-=N_+=9/22$.

between ground-state sublevels g_-, g_+ that does not change the atom energy, $(\omega \pm kv)-(\omega \mp kv)=0$. The energy conservation law thus directly shows that two-photon resonance structures are located at zero velocity, $kv=0$. The widths of the narrow resonance structures can also be estimated from a simple physical argument. For an atom not perturbed by any external interaction, the decay rate for ground-state coherence is zero. A laser field connects ground-state probability amplitudes with upper-state probability amplitudes through the dipole interaction term $\hbar\Omega$ and accordingly causes the decay rate of the ground-state coherence to be of the order of the rate of dipole transitions, i.e., of the order of $\gamma\Omega^2/(\gamma^2+\delta^2)$. This quantity plays accordingly the role of the width μ for narrow resonance structures. Finally the sign of the ground-state coherence at small velocities according to Eqs. (2) and (3) is positive while for a pure Λ scheme the sign can be shown to be negative at small velocities. The last argument explains the existence of the constructive interference in a VVV-interaction scheme contrary to destructive interference in a Λ scheme.

The existence of the narrow two-photon resonance structures in V···V-interaction schemes reflects on any quantities related to the processes of photon absorption and emission. One of the important quantities is the dipole radiation force $\mathbf{F}=\boldsymbol{\nabla}(\langle\mathbf{D}\rangle\mathbf{E})$, where $\langle\mathbf{D}\rangle$ is an induced dipole moment of an atom, and the gradient is assumed to act on the electric field $\mathbf{E}$ only [8]. In the above low-intensity limit, the radiation force is determined by an expression $\mathbf{F}=F\mathbf{e}_z$,

$$F=\hbar k\gamma\Bigg[(2s_- - s_+/3)N_+ + (s_- - s_+)N_0 - (2s_+ - s_-/3)N_- + \frac{1}{6}(s_- - s_+)(\nu+\nu^*) - \frac{i}{6\gamma}(s_-\delta_- + s_+\delta_+)(\nu-\nu^*)\Bigg]. \tag{5}$$

After substitution the ground-state populations and the ground-state coherence, Eq. (5) gives the final expression for the radiation force in an important region of small velocities $kv \ll \gamma$,

$$F=\hbar k\gamma\frac{G}{(1+\delta^2/\gamma^2)}\frac{1}{\Delta}\left[25+\frac{\mu^2}{\mu^2+k^2v^2}\left(\frac{24}{5}\frac{\delta^2}{\gamma^2}-1\right)\right]\times\frac{\delta kv}{\delta^2+\gamma^2}+\hbar k\gamma\frac{G^2}{(1+\delta^2/\gamma^2)^2}\frac{5}{4\Delta}\frac{\delta kv}{\mu^2+k^2v^2}, \tag{6}$$

where $G=2\Omega^2/\gamma^2$ is a dimensionless saturation parameter.

Considering the last expression for the radiation force jointly with expressions for ground-state populations and coherence, one can see that in the low-intensity limit and in the low-velocity region the radiation force includes two parts (Fig. 4). The first part of the force is due to one-photon absorption (emission) processes slightly perturbed by two-photon coherent processes. This incoherent part of the force, F_{in}, has the same physical origin as the force on a two-level atom in the field of two counterpropagating waves [9]. The velocity dependence of the incoherent part of the force is broad since one-photon resonances are centered at resonance velocities $kv=\pm\delta$. The second coherent part of the force,

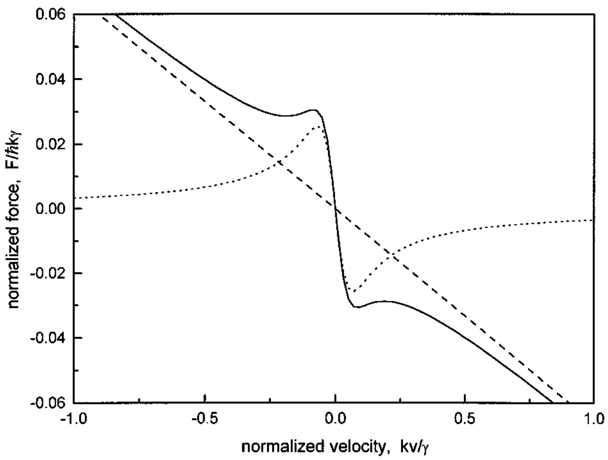

FIG. 4. Radiation force F (solid line), the incoherent part F_{in} of the force (dashed line), and coherent part F_c of the force (dotted line) as functions of velocity for the same parameters as in Fig. 2.

F_c, that is proportional to the square of the laser field intensity is due to the two-photon resonance processes located at zero velocity.

It is to be stressed that at a negative detuning, both incoherent and coherent parts of the force reduce to cooling forces [10]. The friction due to the coherent part of the force is, however, much higher, because the velocity width of the two-photon structure is much less than that due to the one-photon processes. The high friction coefficient in the σ^+-σ^- laser field configuration caused by the radiation force was early on attributed to the gradient of the laser field polarization and the cooling process was described as ''polarization gradient cooling'' [10]. The above results derived for the case of a (3+5)-level atom show that a high friction coefficient in the σ^+-σ^- laser field configuration originates from the two-photon resonance processes that are thus responsible for the deep laser cooling of atoms. Since more complicated V$\cdots$V-interaction schemes include the simplest (3+5)-level scheme, the above statement remains generally valid. To be sure of this point we have also calculated the atomic populations and the radiation pressure force for a (4+6)-level scheme and a (5+7)-level scheme. In all cases we have found that two-photon and higher-order multiphoton processes are responsible for the coherent redistribution of atomic populations at low velocities and the high friction produced by the radiation pressure force. The higher-order multiphoton contributions are, however, typically small compared with that of the two-photon processes. By that reason basic evaluations of the narrow multiphoton velocity structures for real multilevel interaction schemes can be based on the simplest (3+5)-level scheme.

We conclude that the ground-state coherence caused by two-photon processes plays an essential role in optical excitation of multilevel atoms. Independent of the initial internal state of an atom, the ground-state coherence is always produced by two-photon interaction processes. Two-photon processes cause an enhanced atomic population in the upper-state sublevels, produce narrow velocity structures in the ground-state sublevels, and enhance the radiation force at small velocities, leading to a two-photon laser cooling mechanism in the multilevel interaction schemes.

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