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Coherent quantum engineering of free-space laser cooling

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Two distinct lasers are shown to permit controlled cooling of a three-level atomic system to a regime particularly useful for group-II atoms. Alkaline-earth-metal atoms are difficult to laser cool to the micro- or nanokelvin regime, but this technique exhibits encouraging potential to circumvent current roadblocks. Introduction of a sparse-matrix technique permits efficient solution of the master equation for the stationary density matrix, including the quantized atomic momentum. This overcomes long-standing inefficiencies of exact solution methods, and it sidesteps inaccuracies of frequently implemented semiclassical approximations. The realistic theoretical limiting temperatures are optimized over the full parameter space of detunings and intensities. A qualitative interpretation based on the phenomenon of electromagnetically induced transparency reveals dynamical effects due to photon-atom dressing interactions that generate non-Lorentzian line shapes. Through coherent engineering of an asymmetric Fano-type profile, the temperature can be lowered down to the recoil limit range.

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The inability to cool alkaline-earth-metal atoms down to the 100 nK range has proven to be a formidable bottleneck in the study of ultracold dilute gases. Novel cooling techniques have limited applicability, and so far quantum degeneracy has been obtained only for ytterbium, a lanthanide atom with similar level structure, despite extensive efforts by numerous groups [1–4]. The theoretical description of laser cooling took strides in the 1990s, primarily utilizing the semiclassical approximation which resulted in complicated calculations [5]. Few realistic calculations have treated the atomic motion quantum mechanically or predicted realistic cooling temperatures. Here we introduce a direct method based on sparse-matrix techniques, which efficiently yields the fully quantized stationary density matrix. In contrast to Monte Carlo approaches [4,6,7], the temperature and other observables emerge without statistical monitoring; the efficiency enables a complete mapping of the large parameter space of laser intensities and detunings. Here we compute this mapping for a three-level cooling scheme and develop a qualitative interpretation based on the physics of electromagnetically induced transparency (EIT). The calculations predict that promising regimes exist that should produce record low temperatures and overcome the difficulty of alkaline-earth-metal atom cooling. This is orders of magnitude cooler than could be obtained using the main resonance line alone. Moreover, the cooling can be controlled and adjusted in real time, which should make this method competitive with or superior to other proposed ways of cooling alkaline-earth-metal atoms.

Although leading to many advances [8–10], internal atomic structure greatly restricts the species for which laser cooling can be applied successfully, and determines whether ultracold temperatures—on the order of a few microKelvin—can be achieved. Basic Doppler cooling [11,12] has a minimum temperature proportional to the width of the atomic transition. Lower temperatures can be achieved by exploiting the multilevel hyperfine structure [13–16], by modifying the atomic scattering [17], or by using narrow optical transitions as a second-stage for precooled atoms [1,2]. Here, we explore sub-Doppler cooling with three-level systems under two-color excitation. The published literature discusses some similar schemes, for both atoms [18–20] and trapped ions [21–23]. In particular, the work of Tan *et al.* [19] appears initially to be the most exciting, since it claims to permit cooling to below the recoil temperature, believed to be a rigorous quantum lower bound for this type of laser cooling. However, this is an artifact of the errors associated with a semiclassical treatment of atomic momentum in Ref. [19], since that approximation is known to produce incorrect, arbitrarily low (and even negative) temperatures. In order to realistically evaluate the promise of three-level cooling based on dressed-state ideas, it is thus critical to carry out fully quantum solutions with great efficiency, as in the sparse solution technique introduced here.

There are three basic types of three-level systems—the Λ, V, and Ξ systems—each classified according to the ordering of the bare quantum states in energy, and the possible decay pathways. The Λ configuration is commonly used for studying EIT [24] and related phenomena. Here we focus on a Ξ system, shown in Fig. 1, because of its relevance to current experimental work [25,26]. We note, however, that our general conclusions can be applied to any type of three-level system, and can be extended to systems with more than three levels. The level structure and internal parameters for the Ξ system are depicted in Fig. 1. This cooling technique seems well suited to alkaline-earth-metal atoms, which are good candidates for the next generation of optical atomic clocks, studies of ultracold collisions, optical Feshbach resonances [27], and quantum degeneracy [28].

Figure 1 shows the internal atomic states in order of increasing energy as $|0\rangle$, $|1\rangle$, and $|2\rangle$. The transition energy of the lower transition $|0\rangle \rightarrow |1\rangle$ and of the upper transition $|1\rangle \rightarrow |2\rangle$ are $\hbar\omega_0^{(1)}$ and $\hbar\omega_0^{(2)}$, respectively. We include two lasers, of frequencies ω_1 and ω_2, and define their detunings from the appropriate atomic transitions as $\delta_i = \omega_i - \omega_0^{(i)}$, for i

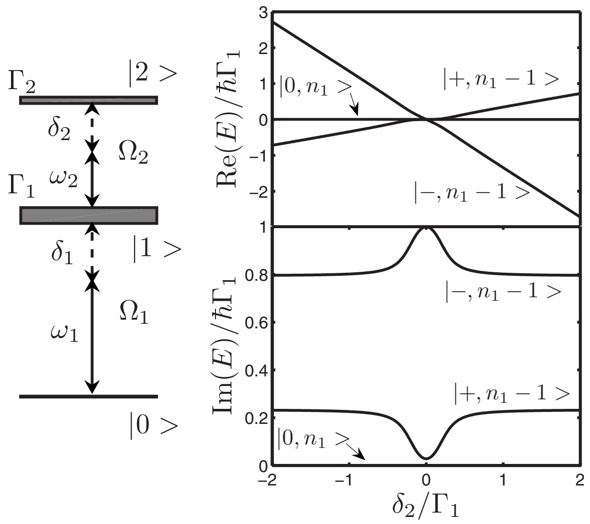

FIG. 1. Left side: Atomic configuration for the Ξ system in ^{24}Mg (see text). Right side: Dressed system, with $s_1(\delta_1)=0.001$ and $s_2(\delta_2)=1$. Real (top) and imaginary (bottom) parts of the eigenvalues of Eq. (3), with dressed atomic states labeled. The real parts are the energies and the imaginary parts are the effective linewidths of the dressed atomic system. Both are plotted as functions of δ_2, with fixed $\delta_1=0$.

$=1,2$. The intensities of lasers 1 and 2 are characterized by their Rabi frequencies $\Omega_1=-\langle 0|\mathbf{d}|1\rangle\cdot\mathbf{E}_1(\mathbf{x})$ and $\Omega_2=-\langle 1|\mathbf{d}|2\rangle\cdot\mathbf{E}_2(\mathbf{x})$, respectively, where $\mathbf{d}$ is the electric-dipole operator of the atom and $\mathbf{E}_i$ is the electric-field amplitude for the ith laser. The spontaneous-emission linewidths of states $|1\rangle$ and $|2\rangle$ are $\Gamma_1/2\pi$ and $\Gamma_2/2\pi$ (79 and 2.2 MHz, respectively, for ^{24}Mg). The time evolution for the atom in the laser field, with mass m and center-of-mass momentum operator $\mathbf{p}$, is given by the master equation

$$\dot{\rho}(t)=i/\hbar[\rho,H]+\mathcal{L}[\rho], \qquad (1)$$

where ρ is the reduced density matrix of the atom system, the vacuum photon field degrees of freedom having been traced over, $H=\mathbf{p}^2/2m+\hbar\omega_0^{(1)}|1\rangle\langle 1|+\hbar\omega_0^{(2)}|2\rangle\langle 2|+V_{\text{laser}}^{(1)}(\mathbf{x})+V_{\text{laser}}^{(2)}\times(\mathbf{x})$ is the Hamiltonian of the atom-laser system, and the superoperator Liouvillian $\mathcal{L}$ describes effects due to coupling of the atom to the vacuum photon field, resulting in spontaneous emission [29].

Equation (1) treats all the atomic degrees of freedom quantum mechanically, so its solutions generally provide an accurate description of the atom's dynamics. We thus avoid much of the difficulty associated with semiclassical approximations of the system. For reasonable temperatures, however, the number of numerical basis states required to directly solve the problem, even in one dimension (1D), is impractical for most computers. Here we resolve this problem without resorting to Monte Carlo methods by noting that the matrix for the linear system equivalent to Eq. (1) is very sparse: only a small fraction of its elements are nonzero. For a typical calculation, the Hilbert space involves three internal degrees of freedom for the atom, and 150 momentum states in 1D. The resulting number of complex density matrix elements is thus $450\times 451/2$ independent elements for this case owing to symmetry, and $\mathcal{L}$ would take over 160 Gbytes to store if it was a full matrix. But the particular structure shared by Liouvillian operators $\mathcal{L}$ that describe relaxation processes simplifies the numerics, and requires less than 2 Gbytes to solve the sparse system; specifically, the microscopic properties of atomic operators in $\mathcal{L}$ permit construction of the matrix $\mathcal{L}$ with the zero elements eliminated. The steady-state solution of Eq. (1) is then found using a standard sparse-matrix inversion, giving an exact direct solution of a fully quantized master equation in just 1–2 min of CPU time on a current workstation.

The steady-state density matrix has been calculated this way in 1D. The average kinetic energy is $\langle p^2/2m\rangle=\mathrm{Tr}(\rho p^2/2m)$, which we equate with $\frac{1}{2}k_BT$, where T is the temperature. The large parameter space of the problem has been explored, e.g., by varying the detunings, δ_1 and δ_2, as well as the strengths Ω_1 and Ω_2 of the two lasers. However, we find that the lasers are better characterized by how strongly they dress the atom. The saturation parameters s_1 and s_2 for the respective transitions are

$$s_i=\frac{1}{2}\frac{\Omega_i^2}{\delta_i^2+(\Gamma_i/2)^2}. \qquad (2)$$

The parameter space has three distinct regimes. In the first, with $s_1\gtrsim 1$ and s_2 arbitrary, only heating occurs, as expected from Doppler-cooling theory since the lower transition is driven strongly. In the second, with $s_1\ll 1$ and $s_2\ll 1$, cooling occurs only to the Doppler limit for the lower transition $T_D^{(1\text{D})}=\hbar\Gamma_1/2k_B$, and only in the range near $\delta_1=-\Gamma_1/2$. In this case, laser 2 has no effect, as this amounts to simple two-level Doppler cooling on the lower transition with laser 1. In the final regime, when $s_1\ll 1$ and $s_2\gtrsim 1$, cooling occurs down to substantially below $T_D^{(1\text{D})}$. Figure 2 illustrates cooling in this regime for ^{24}Mg, with the temperature normalized to $T_D^{(1\text{D})}$, and with the bare two-photon resonance ($\delta_1+\delta_2=0$) denoted by the dashed line. The steady-state temperature is plotted as a function of δ_1 and δ_2 for $s_1(\delta_1)=0.001$ and $s_2(\delta_2)=1$. Note that the saturation parameters are being held fixed as the detunings are varied, so that the Rabi frequencies are being continuously adjusted. We see the lowest temperatures, on the order of $10^{-2}T_D^{(1\text{D})}$, in the quadrant with $\delta_1>0$ and $\delta_2<0$, as well as less extreme cooling in other regions. Observe that the lowest temperatures are obtained for frequencies detuned to the blue of the two-photon resonance. This seems counterintuitive, since a red detuning is usually required in order to have a net decrease of atomic kinetic energy in a photon-scattering event.

Qualitative understanding of the cooling mechanism emerges from analysis of the simpler Hamiltonian

$$H=\frac{\hbar}{2}\begin{pmatrix}0 & \Omega_1 & 0\\ \Omega_1^* & -2\delta_1-i\Gamma_1 & \Omega_2\\ 0 & \Omega_2^* & -2(\delta_1+\delta_2)-i\Gamma_2\end{pmatrix}. \qquad (3)$$

Its complex eigenvalues have real dressed energies, and imaginary parts giving dressed-state linewidths. These dressed energies and widths are plotted on the right side of

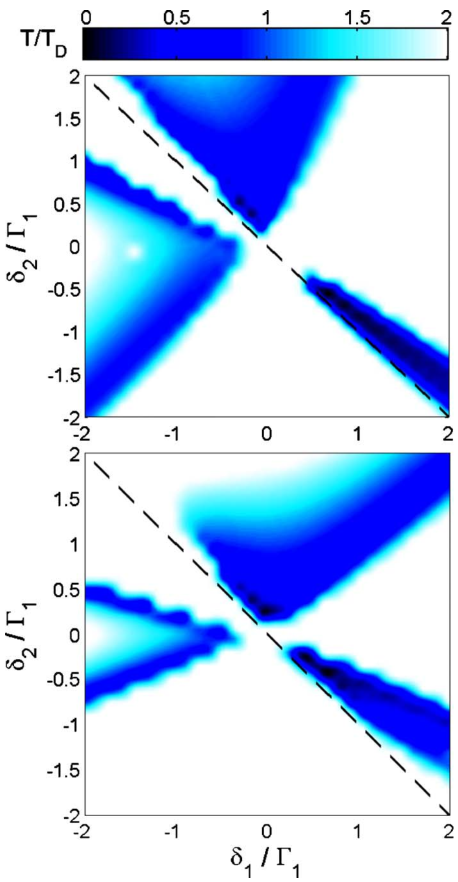

FIG. 2. (Color online) Laser-cooling temperatures for a ^{24}MgΞ system, as a function of δ_1 and δ_2, obtained from exact numerical solutions of Eq. (1). The saturation parameter for the lower transition (probe) is $s_1(\delta_1)=0.001$ for both plots, and for the upper transition (pump) is $s_2(\delta_2)=1$ and 5 in the upper and lower plots, respectively. The temperature is normalized to the 1D Doppler limit for the lower transition $T_D^{(1D)}=7\hbar\Gamma_1/40k_B$ [13], which is the optimum temperature expected for cooling with just one laser. The dashed line indicates the location of the bare two-photon resonance.

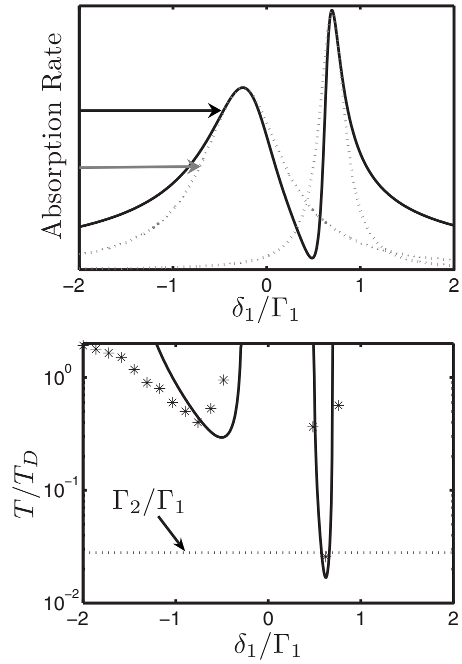

FIG. 3. Upper plot: Absorption spectrum (solid line) as a function of δ_1, illustrating the asymmetric line shapes for the dressed system, for fixed $\delta_2=-\Gamma_1$. The peak of each line shape is located at a dressed eigenenergy, and line shapes at the same energies and widths, but with Lorentzian (symmetric) line shapes are plotted with dotted lines. As an example, optimum-cooling detunings relative to the leftmost resonance are illustrated with a black arrow for the true asymmetric line shape and with a gray arrow for the hypothetical symmetric-line-shape case. The value of the optimum detuning, as well as the slope of the line shape, is seen to be different for these two cases. Lower plot: Solid line, the ratio of the maximum slope of a Lorentzian line shape with width Γ_1 to the slope of the asymmetric line shape, as a function of δ_1 with $\delta_2=-\Gamma_1/2$. This ratio provides an indication of the expected cooling for the dressed system relative to the Doppler limit for the lower transition. For comparison, fully quantum numerical results are indicated by data points.

Fig. 1 as functions of δ_2, for the same parameters used in Fig. 2. The cancellation of one of the widths can be viewed as an EIT effect: since laser 1 is perturbative while laser 2 strongly dresses the upper transition, the new eigenstates of the system, denoted $|+\rangle$ and $|-\rangle$, are well approximated as linear combinations of the bare states $|1\rangle$ and $|2\rangle$. These states have the modified energies and widths shown in Fig. 1. The linewidth modifications can be viewed as a Fano interference [30], in which the dressing-laser transitions caused by the probe laser enable multiple coherent pathways among the bare states. Constructive or destructive interference respectively increases or decreases the atomic linewidth.

The cooling mechanism is thus qualitatively explained as ordinary two-level Doppler cooling. But instead of using a transition between two bare states of an atom, the transition occurs between a (mostly) unmodified ground state, and a dressed excited state, with a shifted energy and a new linewidth that can be narrower than the bare linewidth of the lower transition. As the probe laser is scanned, the detuning relative to the dressed energy levels is varied, but since these levels are shifted from their bare energies, resonance occurs for different detunings than are encountered in the bare system. In fact, the shifts of the eigenenergies in Fig. 1 from the bare energies explain the apparent observation of blue two-photon cooling in Fig. 2. In the dressed system, the bare two-photon resonance is no longer meaningful, and the cooling region is in fact *to the red* of a dressed resonance.

When mapping this system onto Doppler-cooling theory, note that the line shapes are not Lorentzian, but are asymmetric Fano line shapes for the dressed system, as shown in the upper part of Fig. 3 as a function of δ_1 for a fixed value of $\delta_2=-\Gamma_1/2$ [20,31]. This changes the optimum-detuning condition to be that the maximum cooling for a given transition occurs when the probe laser is detuned from the dressed excited state *precisely to the inflection point of the absorption spectrum*. This can be understood by noting that the applied force f due to the laser beam is proportional to the absorption rate, for a given δ_1. As in semiclassical cooling theories, the friction coefficient α for the atom in the laser field is

$$\alpha = -\frac{d}{dv}f(v), \tag{4}$$

where v is the atomic velocity. Since the detuning of the laser and the resonant atomic velocity are linearly related, the derivative of the absorption spectrum with respect to δ_1 also yields a maximum in the cooling force. This is evident in normal Doppler cooling because the optimum detuning occurs when $\delta = -\Gamma/2$, the inflection point of the Lorentzian. In general then, for asymmetric line shapes, the optimum detuning does not obey such a simple relation, but depends on the degree of asymmetry.

From this complete picture of the cooling mechanism, the minimum temperatures can now be determined, allowing for the detuning modification due to asymmetric line shapes. The lower part of Fig. 3 shows the ratio of the maximum slope of a Lorentzian line shape with width Γ_1 to the slope of the asymmetric line shape, as a function of δ_1 with $\delta_2 = -\Gamma_1/2$. This ratio indicates the expected cooling for the dressed system relative to the Doppler limit for the lower transition. Note that the expected temperature, due to the asymmetric line shape, is predicted to be lower than the upper-transition Doppler limit, indicated by the dotted line in the lower plot of Fig. 3, a prediction supported by the numerical data.

In conclusion, coherent engineering of a three-level system can optimize the effectiveness of two-level Doppler cooling. Dressed states are created with modified linewidths in the range between the smallest and the largest of the two bare linewidths, and the additional effect of asymmetric line shapes can lead to temperatures below the Doppler limit of either bare transition. The ability to tailor the degree of cooling lends this technique additional utility, and may be particularly useful when applied to the notoriously difficult-to-cool alkaline-earth-metal atoms. A dressing scheme can be suited to the characteristics of a particular atom, and real-time adjustment of the cooling properties can allow narrowing of the velocity-capture range as an atomic gas is cooled. Utilizing such coherent effects should lead to simple schemes for cooling far below the typical Doppler limit.

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