Feasibility of Cooling and Trapping Metastable Alkaline-Earth Atoms

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Metastability and long-range interactions of Mg, Ca, and Sr in the lowest-energy metastable $^3P_2$ state are investigated. The calculated lifetimes are 38 min for Mg^*, 118 min for Ca^*, and 17 min for Sr^*, supporting feasibility of cooling and trapping experiments. The quadrupole-quadrupole long-range interactions of two metastable atoms are evaluated for various molecular symmetries. Hund's case (c) 4_g potential possesses a large 100–1000 K potential barrier. Therefore magnetic trap losses can possibly be reduced using cold metastable atoms in a stretched $M = 2$ state. Calculations were performed in the framework of *ab initio* relativistic configuration interaction method coupled with the random-phase approximation.

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Introduction.—This work is motivated by emerging experiments on cooling and trapping of alkaline-earth atoms (see, e.g., Refs. [1–3]). In particular, the long-lived lowest-energy $^3P_2$ state can serve as an effective ground state in such experiments. Recently Katori *et al.* [4] cooled and trapped metastable Sr and proposed a scheme for a continuous atom laser with a possible application in atom lithography. The purpose of this work is to evaluate properties of metastable $nsnp\ ^3P_2$ states for Mg ($n = 3$), Ca ($n = 4$), and Sr ($n = 5$). In particular, we calculated decay rates of the $nsnp\ ^3P_2$ states. The resulting lifetimes are on the order of 10^3–10^4 s supporting the feasibility of the experiments.

Ultracold collision properties, including scattering lengths, are sensitive to long-range atomic interactions. The dominant van der Waals interaction of two atoms in their respective $^3P_2$ states is described in terms of the quadrupole moment of the atomic state. To assist in determining molecular potentials, the atomic quadrupole moments of Mg^*, Ca^*, and Sr^* are also calculated here, and the relevant C_5 coefficients for various molecular symmetries are tabulated. These coefficients are substantially larger compared to those for metastable noble gases [5]. In particular, Hund's case (c) 4_g potential possesses a large 100–1000 K potential barrier. Therefore magnetic-trap losses can possibly be reduced using cold metastable atoms in a stretched $M = 2$ state [6].

Method.—Ab initio relativistic valence configuration-interaction (CI) method coupled with random-phase approximation (RPA) was employed here. A detailed description of this method [7] will be published elsewhere; only a brief discussion is presented here. In this method the wave function is expanded in terms of two-particle basis functions as

$$\Psi(\pi JM) = \sum_{k \geq l} c_{kl} \Phi_{kl}(\pi JM)\,. \tag{1}$$

Here J is the total angular momentum with projection M, and π is the parity of the state Ψ. The weights c_{kl} and energies are found by solving the eigenvalue problem based on the *no-pair* Hamiltonian [8]. The basis functions are defined in the subspace of virtual orbitals

$$\Phi_{kl}(\pi JM) = \eta_{kl} \sum_{m_k m_l} C^{JM}_{j_k m_k, j_l m_l} a^\dagger_{\{n_k \kappa_k m_k\}} a^\dagger_{\{n_l \kappa_l m_l\}} |0_{\rm core}\rangle, \tag{2}$$

where sets $\{n\kappa m\}$ enumerate quantum numbers, $\eta^2_{kl} = 1 - \frac{1}{2}\delta_{n_k n_l}\delta_{\kappa_k \kappa_l}$ is a normalization factor, $a^\dagger$ are creation operators, and the quasivacuum state $|0_{\rm core}\rangle$ corresponds to a closed-shell core. The one-particle orbitals, ϕ_k, are found in the frozen core (V^{N-2}) approximation; i.e., the Dirac-Hartree-Fock (DHF) equations are solved self-consistently for core orbitals, and the virtual orbitals are determined using the resulting DHF potential of the core. The employed set of basis functions in Eq. (2) is essentially complete, and one could interpret the solution of the eigenvalue problem as treating the strong Coulomb repulsion of the two valence electrons to all orders of perturbation theory. This valence-CI method, being exact for He-like systems [9], represents an approximation for the alkaline-earth systems. The core-polarization effects and the Breit interaction are neglected here.

Once the wave functions are determined, the matrix elements of a one-particle operator $Z = \sum_{ij} z_{ij} a^\dagger_i a_j$ are computed by forming products $\langle \Psi_F | Z | \Psi_I \rangle$. The additional RPA approximation [10], describing shielding of an externally applied field by core electrons, constitutes a substitution of the "bare" one-particle matrix elements z_{ij} by "dressed" matrix elements $z^{\rm RPA}_{ij}$. Such an approach sums a certain class of many-body diagrams to all orders of perturbation theory.

The configuration-interaction eigenvalue problem was solved numerically using the B-spline basis set technique [11]. The employed basis set included the partial waves $s_{1/2}$-$h_{11/2}$. For each partial wave, 40 positive-energy basis functions approximated by B splines represented a complete set. An inclusion of the 30 lowest-energy basis functions for each partial wave was found to be sufficient for the goals of this work.

Lifetime of $^3P_2$ states.—Single-photon transitions from the metastable $^3P_2$ state are presented in Fig. 1. It will

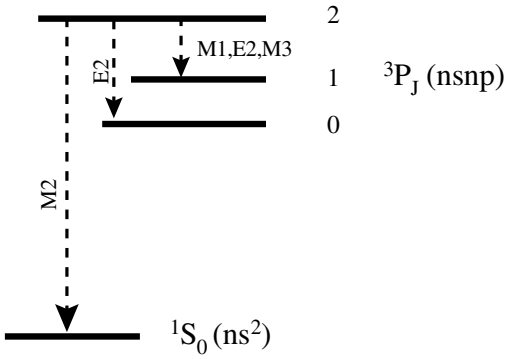

FIG. 1. Single-photon decay channels from the lowest-energy $^3P_2$ state of alkaline-earth atoms.

be demonstrated that for Mg and Ca this level predominantly decays through magnetic-quadrupole ($M2$) transition to the ground $^1S_0$ state. For Sr the main decay channel is a magnetic-dipole ($M1$) transition to the $J = 1$ level of the same ^3P fine-structure multiplet. Estimates show that the magnetic-octupole ($M3$) $^3P_2 \to {}^3P_1$ transition and all second-order two-photon decays contribute a negligibly small fraction to the total decay rate. Further, cold-atom experiments are focused on isotopes without nuclear spin to avoid negotiating molecular potentials complicated by hyperfine structure; we will not consider otherwise important electric-dipole hyperfine-structure-induced decays [12].

The data on magnetic-quadrupole transitions ($M2$) are scarce. For Mg the $^3P_2$-$^1S_0$ $M2$ rates were estimated by Mizushima [13] and Garstang [12] more than three decades ago and recently by Jönsson and Fischer [14]. There are no published data on $M2$ transition rates for Ca and Sr.

A general treatment of multipole electromagnetic transitions in the relativistic framework can be found, for example, in Ref. [9]. The Einstein coefficient A_{M2} for a magnetic-quadrupole transition $I \to F$ is given by

$$A_{M2} = \frac{1}{60}\,\alpha^7\omega^5\,\frac{|\langle\Psi_F||\mathcal{M}^{(2)}||\Psi_I\rangle|^2}{2J_I + 1}, \qquad (3)$$

where ω is the photon frequency and α is the fine-structure constant. (Unless specified otherwise, atomic units $\hbar = |e| = m_e = 1$ are used throughout the paper.) The relevant one-particle reduced matrix element can be expressed as

$$\langle\phi_i||\mathcal{M}^{(2)}||\phi_j\rangle = \frac{2}{3\alpha}\,\langle-\kappa_i||C^{(2)}||\kappa_j\rangle(\kappa_i + \kappa_j) \times \int_0^\infty \left\{15\,\frac{j_2(kr)}{(kr)^2}\right\} r^2[G_i(r)F_j(r) + F_i(r)G_j(r)]\,dr\,. \qquad (4)$$

Here $C^{(2)}$ is the normalized spherical harmonic [15], $G(F)$ are large (small) radial components of a wave function, $\kappa = (l - j)(2j + 1)$, and the photon wave vector is defined as $k = \omega/c$.

The term in curly brackets approaches unity in the long-wavelength approximation. Although the full retarded form of the matrix element was used in the calculations, the long-wavelength limit is well satisfied for transitions between the lowest-energy valence states of alkaline-earth atoms. While the matrix element is largely independent of the transition frequency ω, the rate A_{M2} depends on it strongly ($\sim\omega^5$) and the experimental energy intervals were used in the final tabulation.

The calculated CI+RPA magnetic-quadrupole transition rates are presented in Table I. It was found that the RPA dressing of matrix elements affects these rates only on the order of 1%. The calculated $M2$ transition rate for Mg*, 4.41×10^{-4} s^{-1}, is in a fair agreement with the multiconfiguration Dirac-Fock result, 3.983×10^{-4} s^{-1}, by Jönsson and Fischer [14]. Both rates are twice as large compared to the earlier estimate [12], 1.6×10^{-4} s^{-1}.

Magnetic-dipole $^3P_2 \to {}^3P_1$ transitions occur between the levels of the same fine-structure multiplet. The associated decay rate can be reliably estimated in the nonrelativistic approximation as

$$A_{M1} = \frac{1}{6}\,\alpha^5\omega^3. \qquad (5)$$

The $M1$ rates, presented in Table I, were computed using experimental energy intervals. The determined rate for Mg is 3% larger than in calculations [14] where theoretical fine-structure splittings were employed. The discrepancy is accounted for by a difference between theoretical and experimental energies.

We also calculated electric-quadrupole (E2) decay rates to the $^3P_{1,0}$ fine-structure levels. The results listed in Table I show that the contribution of these decay channels is small compared to the $M2$ and $M1$ rates.

From the analysis of Table I one finds that for Mg and Ca the $^3P_2$ state predominantly decays through magnetic-quadrupole ($M2$) transition to the ground $^1S_0$ state. For Sr the main decay channel is a magnetic-dipole ($M1$) transition to the $J = 1$ level of the same ^3P fine-structure multiplet. Overall, the calculated lifetimes for Mg*, Ca*, and Sr* are on the order of 10^3–10^4 sec, thus favoring the usage of these metastable alkaline-earth atoms in cooling and trapping experiments.

Quadrupole-quadrupole long-range interactions.— Ultracold collision properties, including scattering lengths,

TABLE I. Einstein coefficients A for decays from the lowest-energy $^3P_2$ states in s^{-1}. Notation $x[y]$ stands for $x \times 10^y$.

Type, final state	Mg	Ca	Sr
$M2, ns^2\,{}^1S_0$	4.41[−4]	1.25[−4]	1.27[−4]
$M1, nsnp\,{}^3P_1$	9.12[−7]	1.60[−5]	8.26[−4]
$E2, nsnp\,{}^3P_1$	1[−12]	3[−10]	3[−7]
$E2, nsnp\,{}^3P_0$	3[−12]	1[−9]	1[−6]
A, total	4.42[−4]	1.41[−4]	9.55[−4]

are sensitive to long-range atomic interactions. Here we focus on the long-range interactions of two alkaline-earth atoms in their respective $^3P_2$ states. At large internuclear separations, R, atomic wave functions are perturbed by the axially symmetric molecular potential, which itself depends on the electronic coordinates and the internuclear distance. This potential can be expanded in a sum of interactions of various atomic multipoles [16]. The lowest-order correction to molecular term energies correlating to two $^3P_2$ atomic states arises from a quadrupole-quadrupole contribution [17]

$$V_{QQ} = \frac{1}{R^5} \sum_{\mu=-2}^{2} \frac{4!}{(2-\mu)!\,(2+\mu)!} (Q_\mu)_{\mathrm{I}} (Q_{-\mu})_{\mathrm{II}} . \quad (6)$$

Here subscripts I and II denote the subspaces of the electronic coordinates of the two atoms, and the quadrupole spherical tensor is defined as

$$Q_\mu = -|e| \sum_i r_i^2 C_\mu^{(2)}(\hat{\mathbf{r}}_i) , \quad (7)$$

where the summation is over atomic electrons, $\mathbf{r}_i$ is the position vector of electron i, and $C_\mu^{(2)}(\hat{\mathbf{r}}_i)$ are normalized spherical harmonics [15].

The quadrupole-quadrupole interactions are parametrized by the quadrupole moment $\mathcal{Q}$ of the $^3P_2$ atomic state, defined conventionally as

$$\mathcal{Q} = 2\langle {}^3P_2, M_J = 2 | Q_0 | {}^3P_2, M_J = 2 \rangle . \quad (8)$$

This quadrupole moment is related to the reduced matrix element of the tensor, Eq. (7), as $\mathcal{Q}(^3P_2) = \sqrt{\frac{8}{35}} \langle {}^3P_2 || Q || {}^3P_2 \rangle$. The associated one-particle reduced matrix element is given by

$$\langle \phi_i || Q || \phi_j \rangle = \langle \kappa_i || C^{(2)} || \kappa_j \rangle \times \int_0^\infty r^2 [G_i(r) G_j(r) + F_i(r) F_j(r)]\, dr . \quad (9)$$

Using the valence CI method coupled with the RPA dressing of matrix elements we calculated the quadrupole moments (see Table II). It was found that the RPA sequence of diagrams reduces the final result only by 0.1% for Mg, 0.3% for Ca, and 0.5% for Sr. Because of the diffuse nature of valence states, these quadrupole moments are significantly larger than those found for metastable noble-gas atoms [5], where the hole in the outer $p_{3/2}$ subshell determines the quadrupole moment.

As in the case of metastable $^3P_2$ noble-gas atoms [5,18], the long-range molecular potentials of metastable alkaline-earth atoms depend on their spatial orientation; i.e., the interactions are anisotropic. Altogether, 15 distinct molecular states correlate to the two atomic $^3P_2$ states at large separations. The first order correction to a molecular term may be represented as

$$U^{(1)}(R) = \frac{C_5}{R^5} . \quad (10)$$

The relevant constants C_5 for various molecular symmetries are given in Table III, where the states are characterized using Hund's case (c). These coefficients were obtained by diagonalizing the quadrupole-quadrupole interaction, Eq. (6), in the basis of products of atomic states $|{}^3P_2, M\rangle_{\mathrm{I}} \otimes |{}^3P_2, M'\rangle_{\mathrm{II}}$ for a given symmetry Ω satisfying $M + M' = \Omega$.

Seven of the resulting quadrupole-quadrupole long-range potentials are attractive at large distances and eight are repulsive. We present the most repulsive 0_g^+ and the most attractive 3_g potentials in Fig. 2. It is worth discussing the repulsive 4_g potential also shown in Fig. 2. Provided the metastable atoms in a magnetic trap are prepared in a stretched $M = 2$ state, the collisions would occur along this repulsive $\Omega = 4$ potential. Although the medium-range part of the potential is attractive, the resulting barrier will be on the order of 100–1000 K high. Therefore ultracold collisions of the $^3P_2$ alkaline-earth atoms in a stretched $M = 2$ state can be effectively shielded from the losses occurring at small internuclear distances [6]. This barrier is more pronounced compared to metastable noble-gas atoms [5], where the potential barriers were a few nK high. Clearly, calculations of the second-order C_6 dispersion coefficients and intermediate- and short-range parts of the potentials are needed for a more quantitative description.

Conclusion.—To address the needs of emerging experiments on cooling and trapping of alkaline-earth atoms, we performed *ab initio* relativistic calculations of lifetimes and quadrupole moments of metastable Mg, Ca, and Sr. The determined lifetimes are 38 min for Mg*, 118 min for Ca*, and 17 min for Sr*, supporting the feasibility of experiments. In addition, we investigated long-range quadrupole-quadrupole interactions for molecular potentials correlating to two metastable atoms. Several resulting

TABLE II. Quadrupole moments $\mathcal{Q}$ of the lowest-energy metastable $^3P_2$ states in a.u.

Mg	Ca	Sr
8.59	13.6	16.4

TABLE III. C_5 coefficients in units of $\mathcal{Q}^2$ for molecular states [Hund's case (c)] asymptotically correlating to two $^3P_2$ state atoms. The quadrupole moments $\mathcal{Q}$ are listed in Table II, and long-range interaction potentials are parametrized as $U(R) = C_5/R^5$.

Ω	$C_5/\mathcal{Q}^2$	Ω	$C_5/\mathcal{Q}^2$
0_g^+	2.853 29	3_g	−2.25
0_u^-	2.427 05	2_u	−1.75
4_g	1.5	2_g	−1.625
0_g^+	1.319 89	1_g	−1.236 02
1_u	1.052 02	0_u^-	−0.927 05
1_g	0.986 02	1_u	−0.802 03
2_g	0.75	0_g^+	−0.423 19
3_u	0.75		

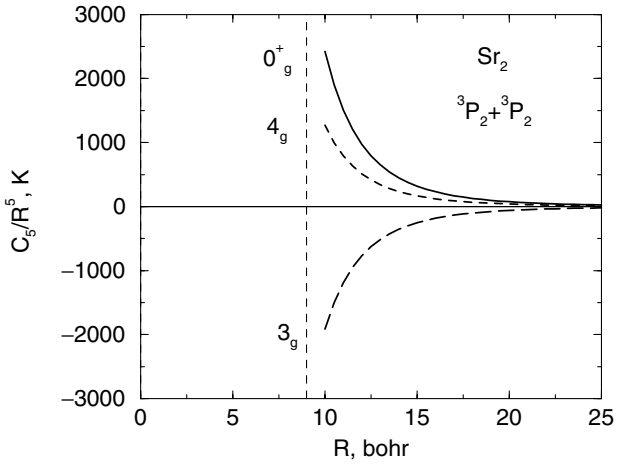

FIG. 2. The most repulsive 0_g^+ and the most attractive 3_g long-range quadrupole-quadrupole interaction potentials correlating to two metastable $5s5p\,^3P_2$ Sr atoms. The 4_g potential is also shown. (1 K = 0.695 03 cm^{-1}.)

potentials possess pronounced barriers, which could be exploited to minimize trap losses.

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