SUPPLEMENTARY MATERIAL

Cesium alignment produced by pumping with unpolarized light

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1 Coefficients for calculating the absorption coefficients $\kappa_0^{F_g \xrightarrow{\text{POL}} F_e}(x)$

In the main text we have claimed that in case of isotropic relaxation one can express the normalized resonant absorption coefficients by algebraic expressions of the form

$$\frac{\kappa_0^{F_g \xrightarrow{\text{POL}} F_e}(x)}{\kappa_{\text{tot}}^{\text{unpol.}}} = \mathcal{D}\,\frac{\sum_n \mathcal{E}_n x^n}{\sum_n \mathcal{F}_n x^n}\,, \tag{S.1}$$

where

$$x = \frac{P_{\text{in}}}{P_{\text{sat}}}. \tag{S.2}$$

The tables below give the numerical values of the parameters $\mathcal{D}$, $\mathcal{E}_n$, and $\mathcal{F}_n$ in Eq. (S.1) for unpolarized (UPL), linearly-polarized (LPL), and circularly-polarized (CPL) light.

Unpolarized light (UPL)

$F_g \to F_e$	4→3	4→4	3→3	3→4
$\mathcal{D}$	189/2	81/2	1	63/2
$\mathcal{E}_0$	231 928 233 984	31 310 311 587 840	33 822 867 456	14 495 514 624
$\mathcal{E}_1$	57 969 475 584	7 525 550 260 224	3 939 762 176	3 623 092 224
$\mathcal{E}_2$	5 400 903 680	641 232 027 648	141 908 992	298 939 392
$\mathcal{E}_3$	220 869 040	22 518 739 152	1 505 903	8 139 835
$\mathcal{E}_4$	3 316 663	263 356 135	0	0
$\mathcal{F}_0$	66 795 331 387 392	5 410 421 842 378 752	154 618 822 656	1 391 569 403 904
$\mathcal{F}_1$	18 757 195 923 456	1 566 298 337 181 696	23 823 646 720	419 765 944 320
$\mathcal{F}_2$	2 053 930 745 856	173 247 078 334 464	1 302 528 000	46 716 092 416
$\mathcal{F}_3$	108 023 480 320	9 042 908 995 584	29 222 400	2 271 631 360
$\mathcal{F}_4$	2 659 739 600	217 778 281 392	215 129	40 699 175
$\mathcal{F}_5$	23 216 641	1 843 492 945	0	0
$\frac{\kappa_0^{F_g \xrightarrow{\text{UPL}} F_e}(0)}{\kappa_{\text{tot}}^{\text{unpol.}}} = \mathcal{D}\frac{\mathcal{E}_0}{\mathcal{F}_0}$	$\frac{21}{64}$	$\frac{15}{64}$	$\frac{7}{64}$	$\frac{21}{64}$

Linearly-plarized light (LPL)

$F_g \to F_e$	4→3 and 3→4	4→4	3→3
$\mathcal{D}$	63/2	9/2	2
$\mathcal{E}_0$	113 246 208	2 548 039 680	8 257 536
$\mathcal{E}_1$	36 691 968	409 300 992	560 896
$\mathcal{E}_2$	3 801 312	17 149 584	6 285
$\mathcal{E}_3$	126 665	157 807	0
$\mathcal{F}_0$	10 871 635 968	48 922 361 856	150 994 944
$\mathcal{F}_1$	4 072 144 896	11 529 879 552	18 808 832
$\mathcal{F}_2$	544 150 016	866 398 464	612 416
$\mathcal{F}_3$	30 902 200	22 717 516	4 403
$\mathcal{F}_4$	633 325	148 393	0
$\frac{\kappa_0^{F_g \xrightarrow{\text{LPL}} F_e}(0)}{\kappa_{\text{tot}}^{\text{unpol.}}} = \mathcal{D}\frac{\mathcal{E}_0}{\mathcal{F}_0}$	$\frac{21}{64}$	$\frac{15}{64}$	$\frac{7}{64}$

Circularly-polarized light (CPL)

$F_g \to F_e$	4→3 and 3→4	4→4	3→3
$\mathcal{D}$	27/2	9/2	6
$\mathcal{E}_0$	638 385 247 179 767 808	27 701 359 832 979 210 240	12 987 981 103 104
$\mathcal{E}_1$	292 316 161 360 527 360	10 609 236 074 922 246 144	2 153 389 228 032
$\mathcal{E}_2$	52 327 072 625 852 416	1 670 724 488 389 459 968	131 538 616 320
$\mathcal{E}_3$	4 597 661 508 304 896	138 313 453 868 679 168	3 505 459 200
$\mathcal{E}_4$	203 069 848 862 720	6 340 752 700 329 984	34 384 475
$\mathcal{E}_5$	4 073 737 696 000	152 426 277 354 176	0
$\mathcal{E}_6$	26 533 150 875	1 499 454 008 045	0
$\mathcal{F}_0$	26 264 993 026 824 732 672	531 866 108 793 200 836 608	712 483 534 798 848
$\mathcal{F}_1$	13 884 879 126 159 949 824	231 768 043 935 926 059 008	147 197 119 168 512
$\mathcal{F}_2$	2 984 541 575 347 961 856	42 764 525 303 645 601 792	11 977 254 502 400
$\mathcal{F}_3$	333 761 207 418 748 928	4 326 637 171 780 878 336	479 258 214 400
$\mathcal{F}_4$	20 586 484 080 312 320	258 937 441 475 493 888	9 422 160 000
$\mathcal{F}_5$	681 137 334 886 400	9 156 177 937 095 680	72 804 375
$\mathcal{F}_6$	10 703 429 268 000	176 931 407 260 608	0
$\mathcal{F}_7$	56 856 751 875	1 440 130 947 255	0
$\frac{\kappa_0^{F_g \xrightarrow{\text{CPL}} F_e}(0)}{\kappa_{\text{tot}}^{\text{unpol.}}} = \mathcal{D}\frac{\mathcal{E}_0}{\mathcal{F}_0}$	$\frac{21}{64}$	$\frac{15}{64}$	$\frac{7}{64}$

2 Coefficients for calculating the orientation $m_{1,0}^{F_g \xrightarrow{\text{POL}} F_e}(x)$

Optical pumping with circularly-polarized light (CPL) can create Cs spin orientation which is characterized by the $m_{k=1,q=0}$ multipole moment of the hyperfine state F_g, defined as

$$m_{1,0}^{F_g \xrightarrow{\text{POL}} F_e}(x) = \sqrt{\frac{3}{F_g(F_g+1)(2F_g+1)}} \sum_{m_g=-F_g}^{F_g} m_g \, p_{F_g,m_g}^{F_g \xrightarrow{\text{POL}} F_e}(x) \,. \tag{S.3}$$

When inserting the algebraic expressions (S.1) for the x-dependence of the sublevel populations, one obtains algebraic expressions of the form

$$m_{1,0}^{F_g \xrightarrow{\text{POL}} F_e}(x) = \mathcal{H} \, \frac{\sum_n \mathcal{I}_n x^n}{\sum_n \mathcal{J}_n x^n} \tag{S.4}$$

for the x-dependence of the orientation. The following table lists the numerical values of the parameters $\mathcal{H}$, $\mathcal{I}_n$, and $\mathcal{J}_n$ for the different hyperfine transitions.

Circularly-polarized light (CPL)

$F_g \to F_e$	4→3	4→4	3→3	3→4
$\mathcal{H}$	$\frac{3}{8}\sqrt{\frac{3}{5}}$	$\frac{1}{2\sqrt{15}}$	$\frac{1}{32\sqrt{7}}$	$\frac{9}{\sqrt{7}}$
$\mathcal{I}_0$	0	0	0	0
$\mathcal{I}_1$	698 233 864 102 871 040	14 499 930 537 575 055 360	127 715 147 513 856	−32 418 000 833 347 584
$\mathcal{I}_2$	317 110 217 286 352 896	5 364 499 008 372 867 072	20 392 840 265 728	−13 689 873 248 550 912
$\mathcal{I}_3$	56 199 131 806 302 208	814 316 853 147 992 064	1 199 791 800 320	−2 190 144 736 067 584
$\mathcal{I}_4$	4 877 033 322 315 776	64 834 427 976 597 504	30 796 870 400	−163 010 263 941 120
$\mathcal{I}_5$	212 100 341 089 280	2 851 652 844 092 352	291 050 775	−5 491 126 345 600
$\mathcal{I}_6$	4 173 067 056 200	65 609 500 803 028	0	−63 950 702 375
$\mathcal{I}_7$	26 533 150 875	616 250 117 245	0	0
$\mathcal{J}_0$	26 264 993 026 824 732 672	531 866 108 793 200 836 608	712 483 534 798 848	26 264 993 026 824 732 672
$\mathcal{J}_1$	13 884 879 126 159 949 824	231 768 043 935 926 059 008	147 197 119 168 512	13 884 879 126 159 949 824
$\mathcal{J}_2$	2 984 541 575 347 961 856	42 764 525 303 645 601 792	11 977 254 502 400	2 984 541 575 347 961 856
$\mathcal{J}_3$	333 761 207 418 748 928	4 326 637 171 780 878 336	479 258 214 400	333 761 207 418 748 928
$\mathcal{J}_4$	20 586 484 080 312 320	258 937 441 475 493 888	9 422 160 000	20 586 484 080 312 320
$\mathcal{J}_5$	681 137 334 886 400	9 156 177 937 095 680	72 804 375	681 137 334 886 400
$\mathcal{J}_6$	10 703 429 268 000	176 931 407 260 608	0	10 703 429 268 000
$\mathcal{J}_7$	56 856 751 875	1 440 130 947 255	0	56 856 751 875

3 Coefficients for calculating the alignment $m_{2,0}^{F_g \xrightarrow{\text{POL}} F_e}(x)$

Optical pumping with unpolarized (UPL), linearly-polarized (LPL), or circularly-polarized (CPL) light creates Cs spin spin alignment which is characterized by the $m_{k=2,q=0}$ multipole moment of the hyperfine state F_g, defined as

$$m_{2,0}^{F_g \xrightarrow{\text{POL}} F_e}(x) = \sqrt{\frac{5}{(2F_g-1)F_g(F_g+1)(2F_g+1)(2F_g+3)}} \sum_{m_g=-F_g}^{F_g} [3m_g^2 - F_g(F_g+1)]\, p_{F_g,m_g}^{F_g \xrightarrow{\text{POL}} F_e}(x)\,. \tag{S.5}$$

When inserting the algebraic expressions (S.1) for the x-dependence of the sublevel populations, one obtains algebraic expressions of the form

$$m_{2,0}^{F_g \xrightarrow{\text{POL}} F_e}(x) = \mathcal{K}\, \frac{\sum_n \mathcal{L}_n x^n}{\sum_n \mathcal{M}_n x^n}\,. \tag{S.6}$$

for the x-dependence of the alignment. The following table lists the numerical values (integer numbers) of the parameters $\mathcal{K}$, $\mathcal{L}_n$, and $\mathcal{M}_n$ for the different hyperfine transitions and light polarizations.

Unpolarized light (UPL)

$F_g \to F_e$	4→3	4→4	3→3	3→4
$\mathcal{K}$	$24\sqrt{77}$	$\frac{2232}{\sqrt{77}}$	$\frac{104}{\sqrt{21}}$	$8\sqrt{21}$
$\mathcal{L}_0$	0	0	0	0
$\mathcal{L}_1$	−1 245 708 288	78 479 622 144	16 515 072	16 515 072
$\mathcal{L}_2$	−263 307 264	16 403 447 808	1 530 880	3 048 448
$\mathcal{L}_3$	−18 769 520	1 113 989 712	34 757	140 305
$\mathcal{L}_4$	−450 163	24 595 375	0	0
$\mathcal{M}_0$	66 795 331 387 392	5 410 421 842 378 752	154 618 822 656	1 391 569 403 904
$\mathcal{M}_1$	18 757 195 923 456	1 566 298 337 181 696	23 823 646 720	419 765 944 320
$\mathcal{M}_2$	2 053 930 745 856	173 247 078 334 464	1 302 528 000	46 716 092 416
$\mathcal{M}_3$	108 023 480 320	9 042 908 995 584	29 222 400	2 271 631 360
$\mathcal{M}_4$	2 659 739 600	217 778 281 392	215 129	40 699 175
$\mathcal{M}_5$	23 216 641	1 843 492 945	0	0

Linearly-polarized light (LPL)

$F_g \to F_e$	$4\to3$	$4\to4$	$3\to3$	$3\to4$
$\mathcal{K}$	$\sqrt{77}$	$\frac{31}{6\sqrt{77}}$	$\frac{13}{8\sqrt{21}}$	$\sqrt{21}$
$\mathcal{L}_0$	0	0	0	0
$\mathcal{L}_1$	9 732 096	−613 122 048	−2 064 384	−2 064 384
$\mathcal{L}_2$	3 191 232	−79 396 416	−95 296	−529 472
$\mathcal{L}_3$	336 567	−2 304 093	−437	−33 025
$\mathcal{L}_4$	11 515	−9 310	0	0
$\mathcal{M}_0$	10 871 635 968	48 922 361 856	150 994 944	10 871 635 968
$\mathcal{M}_1$	4 072 144 896	11 529 879 552	18 808 832	4 072 144 896
$\mathcal{M}_2$	544 150 016	866 398 464	612 416	544 150 016
$\mathcal{M}_3$	30 902 200	22 717 516	4 403	30 902 200
$\mathcal{M}_4$	633 325	148 393	0	0

Circularly-polarized light (CPL)

$F_g \to F_e$	$4\to3$	$4\to4$	$3\to3$	$3\to4$
$\mathcal{K}$	$\frac{3}{8\sqrt{77}}$	$\frac{1}{6\sqrt{77}}$	$\frac{1}{32\sqrt{21}}$	$\sqrt{\frac{3}{7}}$
$\mathcal{L}_0$	0	0	0	0
$\mathcal{L}_1$	−2 413 894 215 898 497 024	103 317 415 502 064 648 192	253 265 631 510 528	17 455 846 602 571 776
$\mathcal{L}_2$	−891 447 400 319 680 512	38 693 712 011 521 425 408	39 521 416 642 560	10 413 655 015 292 928
$\mathcal{L}_3$	−111 643 090 384 584 704	5 917 295 187 540 836 352	2 242 190 376 960	2 309 945 835 913 216
$\mathcal{L}_4$	−4 457 583 294 545 920	471 898 540 024 381 440	54 690 899 200	237 189 760 122 880
$\mathcal{L}_5$	107 635 271 439 360	20 647 720 979 596 992	485 084 625	11 091 122 627 200
$\mathcal{L}_6$	10 101 821 076 200	468 904 175 781 076	0	182 570 968 125
$\mathcal{L}_7$	132 665 754 375	4 313 750 820 715	0	0
$\mathcal{N}_0$	26 264 993 026 824 732 672	531 866 108 793 200 836 608	712 483 534 798 848	26 264 993 026 824 732 672
$\mathcal{M}_1$	13 884 879 126 159 949 824	231 768 043 935 926 059 008	147 197 119 168 512	13 884 879 126 159 949 824
$\mathcal{M}_2$	2 984 541 575 347 961 856	42 764 525 303 645 601 792	11 977 254 502 400	2 984 541 575 347 961 856
$\mathcal{M}_3$	333 761 207 418 748 928	4 326 637 171 780 878 336	479 258 214 400	333 761 207 418 748 928
$\mathcal{M}_4$	20 586 484 080 312 320	258 937 441 475 493 888	9 422 160 000	20 586 484 080 312 320
$\mathcal{M}_5$	681 137 334 886 400	9 156 177 937 095 680	72 804 375	681 137 334 886 400
$\mathcal{M}_6$	10 703 429 268 000	176 931 407 260 608	0	10 703 429 268 000
$\mathcal{M}_7$	56 856 751 875	1 440 130 947 255	0	56 856 751 875