Effect of radiation trapping on the polarization of an optically pumped alkali-metal vapor in a weak magnetic field

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Calculations are presented on the effect of radiation trapping on the electron-spin polarization that can be produced by optical pumping of an alkali-metal vapor in a weak magnetic field. Radiation trapping severely limits the polarization that can be obtained in a weak magnetic field.

In an earlier paper[1] we examined theoretically the effect of radiation trapping on the polarization of an optically pumped alkali-metal vapor in a magnetic field strong enough that the various Zeeman components of the radiation emitted in a transition from the lowest $^2P_{1/2}$ level to the $^2S_{1/2}$ ground level were completely isolated. Recent experimental results agree well with our theoretical calculations.[2] In this paper, we present the results of similar calculations on an optically pumped alkali-metal vapor in a magnetic field weak enough that the various Zeeman components of the $^2P_{1/2}\rightarrow{}^2S_{1/2}$ resonance transition have wavelengths that fall within the Doppler linewidth of the ground level to excited level absorption so that radiation emitted on any Zeeman transition between an excited state and a ground state can be absorbed by other Zeeman transitions.

We use the same notation, the same state labels, and make assumptions similar to those described in Ref. 1. We analyze the effects of radiation trapping on the optical pumping of an "idealized" alkali-metal atom with an electron spin $S=\frac{1}{2}$ and with zero nuclear spin. The states are labeled as follows: The $^2S_{1/2}$, $m=-\frac{1}{2}$ and $m=\frac{1}{2}$ states are labeled 1 and 2, respectively, and the $^2P_{1/2}$, $m=-\frac{1}{2}$ and $m=\frac{1}{2}$ states are labeled 3 and 4, respectively. We assume that the pumping utilizes σ^- light so that it is absorbed only by the $2\rightarrow3$ transition. We assume that the intensity of the pumping light is the same throughout the alkali-metal vapor, and that it covers the absorption line with constant intensity per unit frequency $I_{\bar{\nu}}$. The alkali-metal vapor is assumed to be confined to an infinitely long cylinder of radius R and with completely absorbing walls. We also assume that the densities of atoms in states 1, 2, 3, and 4 are independent of position in the cylinder. These assumptions are discussed in detail in Ref. 1. Our assumptions tend to exaggerate the effects of radiation trapping somewhat; the results that we obtain with these calculations represent a lower limit to the polarization.

Using these assumptions, we can write the rate equations governing the time evolution of the system. The rate equation for state 2 is the following:

$$\frac{dn_2}{dt}=-\frac{I_{\bar{\nu}}}{h\nu}\frac{\lambda^2}{4\pi}A(n_2-n_3)+n_3A_{32}+n_4A_{42}-\frac{(n_2-n_1)}{T_1}$$
$$-n_3(R_{(3\rightarrow2)2}+R_{(3\rightarrow1)2})$$
$$-n_4(R_{(4\rightarrow2)2}+R_{(4\rightarrow1)2})\,. \tag{1}$$

In the rate equation, n_1, n_2, n_3, and n_4 are, respectively, the densities of atoms in states 1, 2, 3, and 4. The Einstein A coefficients for spontaneous emission from the excited states are A_{31}, A_{32}, A_{41}, A_{42}, and $A=A_{31}+A_{32}=A_{41}+A_{42}$. The ratio of the A coefficients is $A_{32}/A_{31}=A_{42}/A_{41}=2$. The wavelength and frequency of the pumping light are, respectively, λ and ν. The ground-state relaxation time is T_1. The rate equations for states 1, 3, and 4 are given in the Appendix.

The first term in the low-field rate equation for n_2 describes the absorption and induced emission of the pumping light. The next two terms represent the spontaneous emission from states 3 and 4 to state 2. The next term is the ground-level relaxation between states 1 and 2. The remaining terms labeled R are due to the radiation trapping. The subscript in parentheses is the transition from which the radiation originates and the second subscript is the state affected. For example, the first radiation trapping term in the rate equation, $n_3R_{(3\rightarrow2)2}$, represents the rate of change in n_2 due to radiation from spontaneous emission from state 3 to state 2.

The radiation trapping terms are the following:

$$n_3R_{(3\rightarrow2)2}=\int\left[d^3\mathbf{r}'\,d\nu\,n_3A\frac{1+\cos^2\theta}{8\pi|\mathbf{r}-\mathbf{r}'|^2}g(\nu-\nu_0)\right.$$
$$\times\exp\{[-\sigma_{-1}^{+1}(n_2-n_3)-\sigma_0^{+1}(n_2-n_4)-\sigma_0^{+1}(n_1-n_3)-\sigma_{+1}^{+1}(n_1-n_4)]|\mathbf{r}-\mathbf{r}'|\}$$
$$\left.\times[\sigma_0^{+1}(n_2-n_4)+\sigma_{-1}^{+1}(n_2-n_3)]\right]\,, \tag{2}$$

$$n_3 R_{(3\to 1)2} = \int \left[d^3\mathbf{r}'\, d\nu\, n_3 A \frac{\sin^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^0_0(n_2-n_4)+\sigma^0_{-1}(n_2-n_3)] \right] , \tag{3}$$

$$n_4 R_{(4\to 2)2} = \int \left[d^3\mathbf{r}'\, d\nu\, n_4 A \frac{\sin^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^0_0(n_2-n_4)+\sigma^0_{-1}(n_2-n_3)] \right] , \tag{4}$$

$$n_4 R_{(4\to 1)2} = \int \left[d^3\mathbf{r}'\, d\nu\, n_4 A \frac{1+\cos^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^{-1}_{-1}(n_2-n_3)-\sigma^{-1}_0(n_2-n_4)-\sigma^{-1}_0(n_1-n_3)-\sigma^{-1}_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^{-1}_0(n_2-n_4)+\sigma^{-1}_{-1}(n_2-n_3)] \right] , \tag{5}$$

where $\sigma^{\Delta m,\ \text{emission}}_{\Delta m,\ \text{absorption}}$ are

$$\sigma^0_0 = \sigma \sin^2\theta , \tag{6}$$

$$\sigma^0_{+1} = \sigma^0_{-1} = \sigma \cos^2\theta , \tag{7}$$

$$\sigma^{+1}_{+1} = \sigma^{-1}_{-1} = \sigma \frac{\sin^4\theta}{(1+\cos^2\theta)} , \tag{8}$$

$$\sigma^{+1}_{-1} = \sigma^{-1}_{+1} = \sigma(1+\cos^2\theta) , \tag{9}$$

$$\sigma^{+1}_0 = \sigma^{-1}_0 = \sigma \frac{\sin^2\theta\cos^2\theta}{(1+\cos^2\theta)} , \tag{10}$$

and

$$\sigma = \frac{\lambda^2}{8\pi} A g(\nu-\nu_0) . \tag{11}$$

The expression for $n_3 R_{(3\to 2)2}$ has the following interpretation. Atoms in state 3 at position $\mathbf{r}'$ make the transition $3\to 2$, emitting radiation with a Doppler line shape $g(\nu-\nu_0)$ and at an angle θ with respect to the magnetic field. The radiation is attenuated exponentially as it travels from position $\mathbf{r}'$ to position $\mathbf{r}$ due to absorption by the transitions $2\to 3$, $2\to 4$, $1\to 3$, and $1\to 4$. After attenuation, the radiation is absorbed by atoms in states 2 at position $\mathbf{r}$ by the transitions $2\to 3$ and $2\to 4$. The other radiation trapping terms have a similar interpretation. In the rate equation for state 2, we have made use of the assumption that the state populations n_1, n_2, n_3, and n_4 are independent of the positions, so that Eq. (1) of this paper corresponds to Eq. (4) of Ref. 1; a comparison of these equations shows that the rate equations for the optical pumping of an optically thick alkali metal are much more complicated in a weak magnetic field than in a strong magnetic field. Since n_1, n_2, n_3, and n_4 are taken to be independent of position we only need solve for these state populations at one location in order to obtain them at all locations. We solve for the state populations at the center of the long cylindrical volume and we call this location $\mathbf{r}=0$.

The integrations over frequency, angle, and radius in the radiation trapping terms are performed in a manner identical to that in Ref. 1. We calculate the integrals over frequency and angle numerically, using Hermitian and Gaussian integration, respectively. The integration over radius is carried out exactly. Numerical solutions of the rate equations are then calculated using $n_1=n_2=\frac{1}{2}$ and $n_3=n_4=0$ as the initial conditions. This yields the state densities and the electron-spin polarization of the alkali-metal vapor target, $P=(n_2-n_1)/(n_2+n_1)$, as a function of time. We use the following parameters: target temperature $T=600\,K$, cylinder radius $R=7.5\times10^{-3}$ m, alkali-metal atomic mass $m=23$ u, wavelength of the resonance radiation $\lambda=589$ nm, and spontaneous decay rate of the excited state $A=6.1\times10^7\ \text{sec}^{-1}$. These parameters are appropriate for an "idealized" Na target for which the nuclear spin is taken to be 0 rather than $\frac{3}{2}$.

In Fig. 1, we compare the results of these weak magnetic field calculations with the results of previous strong magnetic field calculations. Figure 1 shows the steady-state polarization as a function of the target density $N=n_1+n_2+n_3+n_4$ for a target with $T_1=150\ \mu$s and a

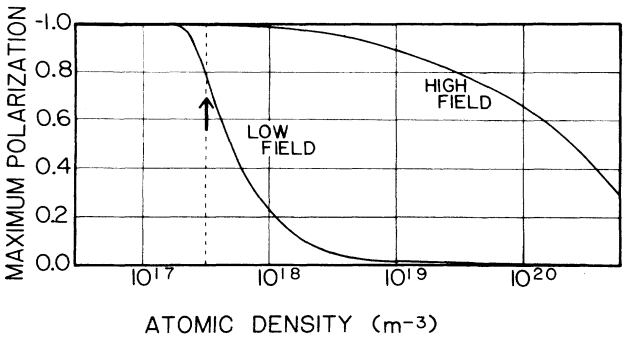

FIG. 1. Steady-state polarizations reached as a function of alkali-metal vapor density, N. The arrow indicates the density for which $(\lambda^2/8\pi)Ag(\nu=\nu_0)NR=1$. We contrast the cases of strong and weak magnetic field. For the calculations we use a cell radius of $R=7.5\times10^{-3}$ m, a ground-state relaxation time of $T_1=150$ μs, and a pumping intensity of $I_{\bar{\nu}}=2.85\times10^{-6}$ W/m^2 Hz, which corresponds to a laser power of 1 W distributed uniformly over the cross-sectional area of the cylinder and over the frequency bandwidth, $g(\nu=\nu_0)^{-1}$, which is nearly the Doppler linewidth.

light intensity per Hz of $I_{\bar{\nu}}=2.85\times10^{-6}$ W/cm^2 Hz. The steady-state polarization obtained by optical pumping in a low magnetic field decreases rapidly for densities greater than $N=1/\sigma_0 R$, where $\sigma_0=(\lambda^2/8\pi)Ag(\nu=\nu_0)$ is the absorption cross section at line center and R is the radius of the cylindrical Na target.

In Table I we present the steady-state polarization as a function of the intensity per unit frequency of the pumping light, total density, and relaxation time. This can be directly compared with Table I of Ref. 1. It is seen that very low polarizations result for the optical pumping in a low magnetic field if the target density is high.

The steady-state solution to the rate equation is not necessarily unique for all initial conditions. For infinite ground-state relaxation time and initial conditions $n_1=N$ and $n_2=n_3=n_4=0$, so that $P=1$, the rate equations indicate that the system remains permanently in this condition. Nevertheless, it is not possible to reach a state with $P=1$ by optical pumping for any other initial condition if radiation trapping is important, even if $T_1=\infty$.

At high density and weak magnetic field, the trapped radiation rapidly removes atoms from states 1 and 2, producing excited states 3 and 4, which decay radiatively to states 1 or 2. Thus the radiation trapping produces a rapid mixing of states 1 and 2 so that a high polarization cannot be obtained if the alkali-metal atomic density is large enough that radiation trapping is important. We find that the polarization of the alkali-metal vapor is almost independent of ground-level relaxation time when radiation trapping is important. This occurs because the net mixing rate of states 1 and 2 is dominated by radiation trapping rather than by relaxation. Thus even a very long relaxation time does not enable one to produce a high polarization under these conditions.

These weak magnetic field results are in marked contrast to the strong magnetic field results. In a strong magnetic field, it is possible to achieve a high polarization in an alkali-metal vapor via optical pumping, provided the radiation trapped decay rate into the highly populated ground state is large compared to the relaxation rate out of this state. Thus in a strong magnetic field, a long ground-level relaxation time is helpful for producing a high polarization via optical pumping.

Radiation trapping acts as an effective mixing of the excited states as well as the ground states. We have also carried out calculations for the case where collisional mixing of the excited states is included in the rate equations. We find that the polarization produced by optical pumping is almost independent of the rate of collisional mixing for the excited states if radiation trapping is important.

Many problems in radiation trapping can be solved by replacing the spontaneous decay rate γ between an excited state and a ground state, with a density-dependent trapped decay rate γ_{eff}. This approach, however, is not satisfactory when the resonant radiation can cause a transition into a second excited state whose population is not proportional to the population of the original excited state. In this case, it is not a single transition that is trapped, and the trapping mixes the two excited states. A simple substitution of γ_{eff} for γ ignores this mixing

TABLE I. Steady-state polarizations reached as a function of the ground-state relaxation time, T_1, and the total atomic density, $N=n_1+n_2+n_3+n_4$. The cases shown are for pumping intensities per unit frequency, $I_{\bar{\nu}}$, of 7.13×10^{-7}, 2.85×10^{-6}, and 1.14×10^{-5} W/m^{-2} Hz. These correspond to laser powers of $\frac{1}{4}$ W, 1 W, and 4 W, respectively, distributed uniformly over the cross-sectional area of the cylinder and over the frequency bandwidth, $g(\nu=\nu_0)^{-1}$, which is nearly the Doppler linewidth. We have used parameters consistent with sodium in a cylindrical target of radius 0.75 cm at a temperature of 600 K.

	$N(m^{-3})$	$\frac{1}{4}$ W	1 W	4 W
$T_1=1.5\times10^{-5}$ s	0	0.894	0.966	0.987
	10^{17}	0.866	0.954	0.980
	10^{18}	0.161	0.219	0.275
	10^{19}	0.003	0.008	0.015
	10^{20}	$<10^{-3}$	$<10^{-3}$	$<10^{-3}$
$T_1=1.5\times10^{-4}$ s	0	0.988	0.996	0.998
	10^{17}	0.972	0.995	0.998
	10^{18}	0.163	0.221	0.276
	10^{19}	0.003	0.008	0.015
	10^{20}	$<10^{-3}$	$<10^{-3}$	$<10^{-3}$
$T_1=1.5\times10^{-3}$ s	0	0.9990	0.9997	0.9998
	10^{17}	0.998	0.9995	0.9998
	10^{18}	0.164	0.221	0.276
	10^{19}	0.003	0.008	0.015
	10^{20}	$<10^{-3}$	$<10^{-3}$	$<10^{-3}$
$T_1=\infty$	0	1.000	1.000	1.000
	10^{17}	1.000	1.000	1.000
	10^{18}	0.164	0.221	0.276
	10^{19}	0.003	0.008	0.015
	10^{20}	$<10^{-3}$	$<10^{-3}$	$<10^{-3}$

and therefore gives an incomplete description of radiation trapping. This is the case for optical pumping of an alkali metal in a weak magnetic field; a photon from any radiative transition may cause an excitation from any ground state to another excited state. On the other hand, for an alkali metal in a strong magnetic field, the Zeeman components of the $^2P_{1/2} \rightarrow {}^2S_{1/2}$ transition are energetically distinct, so that the effect of radiation trapping on the polarization produced in an alkali-metal vapor by optical pumping may be treated with the use of a density- and polarization-dependent effective lifetime.

There is no alkali-metal atom that has nuclear spin 0. The nuclear spin of ^{23}Na is $\frac{3}{2}$. The total angular momentum of the Na atom, **F**, is the vector sum of the electronic angular momentum plus the nuclear spin angular momentum. For both the $3\,^2S_{1/2}$ level and the $3\,^2P_{1/2}$ level, $F=2$ or 1. In the $3\,^2S_{1/2}$ level, the $F=2$ and $F=1$ levels are separated by a hyperfine splitting of 1.77×10^9 Hz, and in the $3\,^2P_{1/2}$ level the $F=2$ and $F=1$ levels are separated by a hyperfine splitting of 1.92×10^8 Hz. As a result of these hyperfine splittings, the optical absorption cross section for light of a given frequency ν is less than it would be if the alkali metal had zero nuclear spin and therefore no hyperfine structure. Thus, radiation trapping produces smaller effects for the real alkali-metal vapor than for a vapor of idealized alkali-metal atoms with $I=0$. It is expected that the polarization obtained by optical pumping at a given density will be somewhat higher than predicted by our calculations.

We have made the assumption of uniform atomic-state densities n_1, n_2, n_3, and n_4 throughout the cylinder. Since we allow the ground-level relaxation time to exceed several flight times across the target, it is expected that the ratio of n_1 and n_2 will not depend greatly on the radial position in the target. However, since radiation may escape the target at the walls, the excited-state density is expected to be less near $r=R$ than at the axis of the cylinder. Our assumption of a spatially flat excited-state density presumes too large an excited state density near the walls of the cylinder. This causes our calculations to overestimate the amount of depolarizing scattered radiation, thereby exaggerating the effects of radiation trapping. Thus, the assumption of uniform atomic-state densities results in an underestimate of the polarization that can be produced by optical pumping.

We conclude that in a weak magnetic field radiation trapping prevents one from obtaining a large polarization in an optically pumped alkali-metal vapor if the target density exceeds $(\sigma_0 R)^{-1}$, even if the ground-level relaxation time is very long. This result has been known experimentally for some time.[3] It is possible to produce a larger polarization by the use of a buffer gas such as N_2 at a pressure high enough to quench the alkali-metal excited states, provided the ground-level relaxation time is large compared to the pumping time.[3] It is also possible to produce a large polarization in a magnetic field that is large enough to cause complete Zeeman separation of the various transitions between excited and ground states, provided the ground-level relaxation time is long compared to the radiatively trapped decay rate into the highly populated state.[1]

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APPENDIX

The rate equations for states 1, 3, and 4 are as follows:

$$\frac{dn_1}{dt} = n_3 A_{31} + n_4 A_{41} + \frac{(n_2-n_1)}{T_1} - n_3(R_{(3\rightarrow1)1}+R_{(3\rightarrow2)1}) - n_4(R_{(4\rightarrow1)1}+R_{(4\rightarrow2)1}) , \tag{A1}$$

$$\frac{dn_3}{dt} = \frac{I_{\bar{\nu}}}{h\nu}\frac{\lambda^2}{4\pi} A (n_2-n_3) - An_3 + n_3(R_{(3\rightarrow1)3}+R_{(3\rightarrow2)3}) + n_4(R_{(4\rightarrow1)3}+R_{(4\rightarrow2)3}) , \tag{A2}$$

$$\frac{dn_4}{dt} = -An_4 + n_3(R_{(3\rightarrow1)4}+R_{(3\rightarrow2)4}) + n_4(R_{(4\rightarrow1)4}+R_{(4\rightarrow2)4}) . \tag{A3}$$

In these equations, the radiation trapping terms are

$$n_3 R_{(3\rightarrow2)1} = \int \left[d^3r'\, d\nu\, n_3 A \frac{1+\cos^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \times \exp\{[-\sigma_{-1}^{+1}(n_2-n_3)-\sigma_0^{+1}(n_2-n_4)-\sigma_0^{+1}(n_1-n_3)-\sigma_{+1}^{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\} \times [\sigma_0^{+1}(n_1-n_3)+\sigma_{+1}^{+1}(n_1-n_4)] \right] , \tag{A4}$$

$$n_3 R_{(3\to 1)1} = \int \left[d^3r'\, d\nu\, n_3\, A \frac{\sin^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^0_0(n_1-n_3)+\sigma^0_{+1}(n_1-n_4)] \right], \tag{A5}$$

$$n_4 R_{(4\to 1)1} = \int \left[d^3r'\, d\nu\, n_4\, A \frac{1+\cos^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^{-1}_{-1}(n_2-n_3)-\sigma_0^{-1}(n_2-n_4)-\sigma_0^{-1}(n_1-n_3)-\sigma^{-1}_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma_0^{-1}(n_1-n_3)+\sigma^{-1}_{+1}(n_1-n_4)] \right], \tag{A6}$$

$$n_4 R_{(4\to 2)1} = \int \left[d^3r'\, d\nu\, n_4\, A \frac{\sin^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^0_0(n_1-n_3)+\sigma^0_{+1}(n_1-n_4)] \right], \tag{A7}$$

$$n_3 R_{(3\to 1)3} = \int \left[d^3r'\, d\nu\, n_3\, A \frac{\sin^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^0_0(n_1-n_3)+\sigma^0_{-1}(n_2-n_3)] \right], \tag{A8}$$

$$n_3 R_{(3\to 2)3} = \int \left[d^3r'\, d\nu\, n_3\, A \frac{1+\cos^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^{+1}_{-1}(n_2-n_3)-\sigma_0^{+1}(n_2-n_4)-\sigma_0^{+1}(n_1-n_3)-\sigma^{+1}_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma_0^{+1}(n_1-n_3)+\sigma^{+1}_{-1}(n_2-n_3)] \right], \tag{A9}$$

$$n_4 R_{(4\to 1)3} = \int \left[d^3r'\, d\nu\, n_4\, A \frac{1+\cos^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^{-1}_{-1}(n_2-n_3)-\sigma_0^{-1}(n_2-n_4)-\sigma_0^{-1}(n_1-n_3)-\sigma^{-1}_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma_0^{-1}(n_1-n_3)+\sigma^{-1}_{-1}(n_2-n_3)] \right], \tag{A10}$$

$$n_4 R_{(4\to 2)3} = \int \left[d^3r'\, d\nu\, n_4\, A \frac{\sin^2\theta}{8\pi\,|\,\mathbf{r}-\mathbf{r}'\,|^2} g(\nu-\nu_0) \right.$$
$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)]\,|\,\mathbf{r}-\mathbf{r}'\,|\}$$
$$\left. \times [\sigma^0_0(n_1-n_3)+\sigma^0_{-1}(n_2-n_3)] \right], \tag{A11}$$

$$n_3 R_{(3\to 1)4} = \int \left[d^3r'\, d\nu\, n_3 A \frac{\sin^2\theta}{8\pi \mid \mathbf{r}-\mathbf{r}' \mid^2} g(\nu-\nu_0) \right.$$

$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)] \mid \mathbf{r}-\mathbf{r}' \mid\}$$

$$\left. \times [\sigma^0_0(n_2-n_4)+\sigma^0_{+1}(n_1-n_4)] \right] , \tag{A12}$$

$$n_3 R_{(3\to 2)4} = \int \left[d^3r'\, d\nu\, n_3 A \frac{1+\cos^2\theta}{8\pi \mid \mathbf{r}-\mathbf{r}' \mid^2} g(\nu-\nu_0) \right.$$

$$\times \exp\{[-\sigma^{+1}_{-1}(n_2-n_3)-\sigma^{+1}_0(n_2-n_4)-\sigma^{+1}_0(n_1-n_3)-\sigma^{+1}_{+1}(n_1-n_4)] \mid \mathbf{r}-\mathbf{r}' \mid\}$$

$$\left. \times [\sigma^{+1}_0(n_2-n_4)+\sigma^{+1}_{+1}(n_1-n_4)] \right] , \tag{A13}$$

$$n_4 R_{(4\to 1)4} = \int \left[d^3r'\, d\nu\, n_4 A \frac{1+\cos^2\theta}{8\pi \mid \mathbf{r}-\mathbf{r}' \mid^2} g(\nu-\nu_0) \right.$$

$$\times \exp\{[-\sigma^{-1}_{-1}(n_2-n_3)-\sigma^{-1}_0(n_2-n_4)-\sigma^{-1}_0(n_1-n_3)-\sigma^{-1}_{+1}(n_1-n_4)] \mid \mathbf{r}-\mathbf{r}' \mid\}$$

$$\left. \times [\sigma^{-1}_0(n_2-n_4)+\sigma^{-1}_{+1}(n_1-n_4)] \right] , \tag{A14}$$

$$n_4 R_{(4\to 2)4} = \int \left[d^3r'\, d\nu\, n_4 A \frac{\sin^2\theta}{8\pi \mid \mathbf{r}-\mathbf{r}' \mid^2} g(\nu-\nu_0) \right.$$

$$\times \exp\{[-\sigma^0_{-1}(n_2-n_3)-\sigma^0_0(n_2-n_4)-\sigma^0_0(n_1-n_3)-\sigma^0_{+1}(n_1-n_4)] \mid \mathbf{r}-\mathbf{r}' \mid\}$$

$$\left. \times [\sigma^0_0(n_2-n_4)+\sigma^0_{+1}(n_1-n_4)] \right] . \tag{A15}$$

For the radiation trapping terms, the cross sections $\sigma^{\Delta m,\ \text{emission}}_{\Delta m,\ \text{absorption}}$ are given in the text, in Eqs. (6)–(11).

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