Optical transmission through a dipolar layer: Supplementary Information

James Keaveney, Ulrich Krohn, Ifan G. Hughes, Armen Sargsyan,
David Sarkisyan, Charles S. Adams

I. SUSCEPTIBILITY AND OPACITY

Using diffraction theory and the superposition principle [1] for a dipolar layer of thickness δz in the $z = 0$ plane, the intensity in a plane a distance z downstream is given by

$$\mathcal{I}_{\mathrm{t}}^{(z)} = \left(1 - \mathrm{Im}[\chi]\frac{\delta z}{\lambda\!\!\!^{-}}\right)\mathcal{I}_0^{(z)} \ , \tag{1}$$

where $\mathcal{I}_{\mathrm{t}}^{(z)}$ and $\mathcal{I}_0^{(z)}$ are the intensities of the transmitted and incident fields, respectively; $\lambda\!\!\!^{-} = \lambda/(2\pi)$ is the reduced wavelength of the transition and χ is the electric susceptibility of the medium. To find the opacity one simply needs an expression for the susceptibility.

For an ideal two-level scatterer with ground state $|g\rangle$ and excited state $|e\rangle$ separated by an energy $\hbar\omega_0$, ignoring atomic motion, the susceptibility in the weak probe limit as a function of the detuning from resonance, $\Delta = \omega - \omega_0$, is expressed as

$$\chi(\Delta) = \frac{\mathrm{i}N\mu_{ge}^2}{\epsilon_0\hbar(\Gamma/2 - i\Delta)} \ , \tag{2}$$

where N is the number density of scatterers, Γ is the full-width at half-maximum (hereafter referred to as the linewidth) of the transition and μ_{ge} is the dipole matrix element of the transition. The opacity (optical depth) of the medium is given by

$$\mathrm{OD} = k\mathrm{Im}[\chi]\delta z = N\sigma\delta z \ . \tag{3}$$

A. No dipole–dipole interactions

Without dipole–dipole interactions, the linewidth of the transition is limited by spontaneous decay, $\Gamma = \Gamma_e$, where the excited state linewidth Γ_e is given by (see [2] and references therein),

$$d_{\mathrm{ge}} = \langle L_g|er|L_e\rangle = \sqrt{3}\sqrt{3\pi\epsilon_0\hbar\Gamma_e\lambda\!\!\!^{-3}} \ , \tag{4}$$

$$\Gamma_e = \frac{d_{\mathrm{ge}}^2}{9\pi\epsilon_0\hbar\lambda\!\!\!^{-3}} \ . \tag{5}$$

For the D_2 line, we have

$$\mu_{ge}^2 = |\langle J_g = 1/2|er|J_e = 3/2\rangle|^2 = \frac{2}{3}|\langle L_g = 0|er|L_e = 1\rangle|^2 = \frac{2}{3}d_{ge}^2 \ , \tag{6}$$

and hence the on-resonance ($\Delta = 0$) susceptibility is given by

$$\chi = \mathrm{i}\frac{3N\lambda\!\!\!^{-3}}{4\pi^2} \ . \tag{7}$$

In this case the incident field and the dipolar field interfere destructively in the forward direction and the transmitted field is attenuated to the value

$$\mathcal{I}_{\mathrm{t}}^{(z)} = \left(1 - \frac{3N\lambda\!\!\!^{-2}}{2\pi}\delta z\right)\mathcal{I}_0^{(z)} \ , \tag{8}$$

which is Beer's law with the resonant optical cross section $\sigma_0 = 6\pi\lambda\!\!\!^{-2}$.

B. Strong dipole-dipole interactions

Resonant dipole–dipole interactions scale as $V_{\rm dd} \propto 1/r^3 \propto N$. The magnitude and sign of the interaction depends on relative orientation of the dipoles, but for an atomic gas the dipoles are randomly distributed, and the interactions manifest as a broadening of atomic resonance lines. The total linewidth becomes

$$\Gamma = \Gamma_e + \Gamma_{\rm dd} = \Gamma_e + \beta N \ , \tag{9}$$

where the coefficient of the broadening, β, is dependent on the transition, and depends on the natural linewidth of the excited state and the transition wavelength [3],

$$\Gamma_{\rm dd} = \beta N = 2\pi G N \Gamma_e \lambda\!\!\!^{-3} \ , \tag{10}$$

where $G = \sqrt{\frac{g_e}{g_g}}$ and the $g_i = 2J_i + 1$ are the degeneracies of the states. Substituting in for the natural linewidth, the self-broadening coefficient is given by

$$\beta = \frac{2}{9}\frac{G d^2}{\hbar \epsilon_0}. \tag{11}$$

As density increases, dipole–dipole interactions become the dominant source of line broadening, and the optical depth of the medium saturates. Combining equations (2) and (10), on resonance ($\Delta = 0$) at high density, the susceptibility reduces to

$$\chi \simeq \mathrm{i}\frac{2\mu_{ge}^2}{\epsilon_0 \hbar \beta} \simeq \mathrm{i}\frac{9\mu_{ge}^2}{G d^2} \ . \tag{12}$$

The detuning, Δ, also becomes relatively unimportant (for the imaginary part of χ) and thus the opacity saturates at a single broadband value. The exact numerical value depends on fine structure, polarisation of the light field and angular averaging of the dipole-dipole interaction. This is equivalent to a resonant optical cross-section that decreases with density, $\sigma_0 = \mathrm{Im}[\chi]/(N\lambda\!\!\!^{-})$.

C. High density saturation on the Rb D_2 line in the 2D limit

As in our case the medium is a gas, the motion of the dipole can change the optical response. Motional effects include transit time broadening, Doppler broadening and collisional dephasing. All of these processes depend on how far the dipole can move along (and across, for transit time broadening) the light propagation direction during its time of emission. For example, if the atom can move less than half the optical wavelength, this suppresses the Doppler effect, leading to the Dicke narrowed resonances we observe at low density in Fig. 3 (see also [4, 5]). Similarly if the atoms move more than $\lambda/4$ this suppresses the dipole–dipole interaction as the motion leads to a phase change of π. For this reason, to study the saturation due to dipole–dipole interactions in a 2D layer we restrict the investigation to cell lengths less than $\lambda/4$.

Including motional effects due to the constrained geometry at thicknesses less than $\lambda/4$, and taking into account the effective reduced dipole moment for linearly polarised light, $d_{\rm lin}^2 = d^2/3$, we substitute in to equation (12) with $G = \sqrt{2}$ and find a maximum opacity of

$$\mathrm{OD} = \frac{1}{\sqrt{2}}\frac{\delta z}{\lambda\!\!\!^{-}}. \tag{13}$$

For a three–dimensional gas, we expect motional effects to be negligible such that $\mathrm{OD} = \sqrt{2}\delta z/\lambda\!\!\!^{-}$.

D. Absorption from a monolayer

For a monolayer of atoms, $\delta z \sim a_0$, the Bohr radius. For simplicity, we take the $1S_{1/2} - 2P_{3/2}$ transition in Hydrogen, which has a wavelength given by the Rydberg formula,

$$\frac{1}{\lambda} = R_\infty\left(\frac{1}{1^2} - \frac{1}{2^2}\right) = \frac{3}{4}R_\infty = \frac{3\alpha^2}{8\lambda_e} = \frac{3}{16}\frac{\alpha}{\pi a_0} \tag{14}$$

$$\frac{1}{\lambda\!\!\!^{-}} = \frac{3}{8}\frac{\alpha}{a_0} \tag{15}$$

with $\lambda_e = 2\pi\alpha a_0$ the Compton wavelength and the Rydberg constant $R_\infty = \alpha^2/2\lambda_e$. In the absence of fine-structure splitting, $G = \sqrt{\frac{2L_e+1}{2L_g+1}} = \sqrt{3}$ and $\mu_{ge} = d$, leading to

$$\sigma = \frac{27}{16\sqrt{3}} \frac{1}{N} \frac{\alpha}{a_0} \sim \frac{1}{N} \frac{\alpha}{a_0} , \tag{16}$$

$$\mathrm{OD} \sim \alpha . \tag{17}$$

II. ADDITIONAL CELL DETAILS

The cell is constructed from two super-polished sapphire windows, of dimensions 20 x 30 mm x 2.5 mm, attached to a reservoir filled with natural abundance Rb. To produce a very small thickness, the inside surface of one of the sapphire plates must have a small curvature of radius $R > 100$ m. Before the final gluing, the sapphire plates are assembled and it is possible to estimate the expected minimum thickness, the range of the thickness variation and the degree of smoothness. For this cell, the local surface roughness measured over an area of 1 mm^2 is less than 3 nm, for any part of the window. There could be a greater total variation across the whole sapphire plate, but since the diameter of the laser at focus is much less than 1 mm^2, this is not important. The windows are glued together, with a 2 μm platinum spacer used to create the wedge-shaped region between the windows, leading to a variable thickness of between ~ 30 nm to 2 μm. The local cell thickness can be determined accurately at operational temperature using an interferometric method outlined in [6].

The reservoir can be heated pseudo-independently of the windows. The reservoir temperature determines the Rb number density, while the windows are kept $\geq 50°$C hotter to prevent condensation of Rb onto the windows.

[1] M. Born and E. Wolf, *Principles of Optics*, 7th ed. (CUP, Cambridge, 1999).

[2] P. Siddons, C. S. Adams, C. Ge, and I. G. Hughes, Journal of Physics B: Atomic, Molecular and Optical Physics **41**, 155004 (Aug. 2008), ISSN 0953-4075, http://stacks.iop.org/0953-4075/41/i=15/a=155004?key=crossref.00c7138be904263053df130d39a795f7.

[3] L. Weller, R. J. Bettles, P. Siddons, C. S. Adams, and I. G. Hughes, Journal of Physics B: Atomic and Molecular Physics **44**, 195006 (2011).

[4] R. Romer and R. Dicke, Physical Review **99**, 532 (Jul. 1955), ISSN 0031-899X, http://link.aps.org/doi/10.1103/PhysRev.99.532.

[5] S. Briaudeau, S. Saltiel, G. Nienhuis, D. Bloch, and M. Ducloy, Physical Review A **57**, R3169 (May 1998), ISSN 1050-2947, http://link.aps.org/doi/10.1103/PhysRevA.57.R3169.

[6] E. Jahier, J. Guéna, P. Jacquier, M. Lintz, A. V. Papoyan, and M. A. Bouchiat, Applied Physics B: Lasers and Optics **71**, 561 (Oct. 2000), ISSN 0946-2171, http://www.springerlink.com/openurl.asp?genre=article&id=doi:10.1007/s003400000388.