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PHOTOPHORESIS AND THE SCATTERING OF ELECTROMAGNETIC RADIATION

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Lawrence Livermore National Laboratory

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PHOTOPHORESIS AND THE SCATTERING OF ELECTROMAGNETIC RADIATION

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1. Effective Dielectric Constant for Soot Particles (with Arthur Broyles)

Electron-microscope photographs of soot lend support to the picture in which a soot particle is modeled as a collection of chains of small carbon spheres. The soot particle itself is typically considerably larger than the small carbon spheres making up the chains. Thus the soot particles might have a size $\sim 0.1 - 1\mu m$ while the small carbon spheres might have a size $\sim 0.03\mu m$ in typical situations. Further, measurements of the density of soot yield values much less than that of normal carbon, indicating that an individual soot particle has a rather small filling factor, i.e., the fraction of the volume of the particle that is occupied by chains.

If a soot particle is taken to be a sphere partially filled with carbon chains, what are its scattering and absorption properties? Several workers have adopted the view that the net scattering and absorption properties can be determined simply by summing the cross-sections for the individual small carbon spheres.

We feel that such a procedure cannot be valid in general because it neglects coherence effects among the various randomly located scatterers within the soot particle.

It appears that in a first rough approximation the scattering and absorption properties of soot can be determined by estimating the effective dielectric constant of a soot sphere. The dielectric constant ϵ of the sphere in the present case will be decreased considerably

below that of a pure carbon sphere of the same size because the soot particle has a filling factor $<< 1$.

To estimate the effective dielectric constant ϵ we use the Clausius-Mossotti relation, which states that, especially for dilute substances,

$$\frac{1}{\rho}\frac{\epsilon - 1}{\epsilon + 2} \simeq \text{ const}, \tag{1}$$

where ρ is the average density of the substance. This relation and the derivation leading to it take into account in a gross way the roles of density and coherent scattering effects in determining the effective value of ϵ. If ρ_o is the density, say, of normal carbon and ρ that of a soot particle, then Eq. (1) implies that

$$\epsilon \simeq \frac{1 + 2\left(\frac{\rho}{\rho_o}\right)\left(\frac{\epsilon_0 - 1}{\epsilon_0 + 2}\right)}{1 - (\rho/\rho_o)\left(\frac{\epsilon_0 - 1}{\epsilon_0 + 2}\right)} \tag{2}$$

where ϵ_0 is the dielectric constant for normal carbon.

As an example, consider soot particle of size $\sim 0.1\mu m$. Such particles have an empirically determined density $\rho \sim 156.2 kg./m^3$. (G. Muholland, private communication). From Eq. (2) and the normal values ρ_0 and ϵ_0 for carbon one obtains

$$n \equiv n_r + i\, n_i \equiv \sqrt{\epsilon} \simeq 1.05 + i\, 3.41 \times 10^{-2} \tag{3}$$

for a $\sim 0.1\mu m$ soot particle. Here n is the effective index of refraction, and n_r and n_i are its real and imaginary parts, respectively. An application of Eq. (3) to the problem of photophoresis will be given in the next section.

2. An Approximate Analysis of the Phenomenon of Photophoresis (with Arthur Broyles)

When electromagnetic radiation passes through a dielective sphere with complex index of refraction (or dielectric constant), the radiation is both attenuated and focused, to some extent, during transit through the sphere. The attenuation favors a situation in which the front hemisphere (that hemisphere facing *towards* the source of the radiation) absorbs more

energy from the radiation field than the back hemispheres (that hemisphere facing *away from* the source of the radiation) does. The focusing, on the other hand, tends to favor a situation in which the back hemisphere absorbs more energy, because the focusing typically strengthens the field in the back hemisphere more so than in the front hemisphere.

In certain situations, these effects cause significantly more energy to be absorbed in the back hemispheres than in the front hemisphere. Due to inelastic interactions with molecules of the surrounding medium, this can lead to the phenomenon of negative, as opposed to positive, photophoresis, in which the dielectric sphere receives a boost in the direction toward, as opposed to away from, the radiation source.

In the next subsection we shall develop an approximate method for calculating the energy absorbed by the front and back hemispheres of dielectric spheres; and in a subsequent subsection we shall apply the method to calculations of the photophoretic effect.

A. The Energy Absorbed in Front and Back Hemispheres

We shall employ an approximate geometrical optics method and later check it by comparing with empirical results.

In geometrical optics, the focal length of a sphere of radius R is

$$f = \frac{R}{2(n_r - 1)}, \tag{4}$$

where n_r is again the real part of the index of refraction.

Consider first the situation in which $f < R$. Then the paths of a bundle of rays impinging on the sphere at an angle of incidence θ is as shown in Figure 1.

The incident beam (plane wave) has Poynting flux (energy per unit area per unit time)

$$S_{\text{in}} = \frac{\tilde{c}}{8\pi}|E_{\text{in}}|^2, \tag{5}$$

where $\tilde{c} \equiv$ speed of light and E_{in} = amplitude of incident electric field, and where we have assumed time dependence e^{-iwt}. Similarly, the energy flux in the annular region shaded,

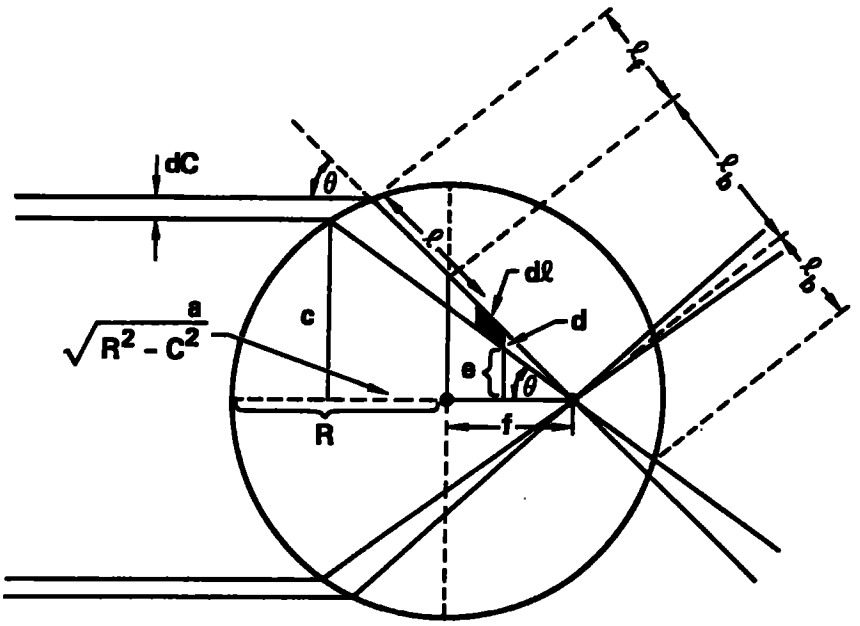

Figure 1. Geometrical-optics approximation for photophoretic effect.

with sides de and dl, is

$$S = \frac{\tilde{c}}{8\pi} n_r |E|^2, \tag{6}$$

where E is the amplitude of the electric field in the shaded region. By conservation of energy, with allowance made for attenuation due to absorption,

$$T|E_{\text{in}}|^2 2ncdc \simeq n_r |E|^2 e^{-2n_i k_o l}\, 2\pi e \cos\theta d\epsilon, \tag{7}$$

where T is the fraction of the incident energy that is transmitted, l is the distance along the beam from the point of entry into the body down to the shaded elemental volume, and k_o is the incident wave number ($k_o = 2\pi/\lambda, \lambda =$ wavelength). Now the rate dR_{abs} of absorption of energy in the annular region of dimensions dl and de is

$$\begin{aligned} dR_{abs} &= \frac{1}{2}\sigma |E|^2 d^3x \\ &= \frac{1}{2}\sigma |E|^2 2\pi e \cos\theta de dl \\ &= \frac{T}{2}\frac{\sigma}{n_r}|E_{\text{in}}|^2 e^{-2n_i k_o l} 2\pi\ c\ d\ c\ d\ l. \end{aligned} \tag{8}$$

Here the conductivity

$$\sigma = \frac{n_r n_i \tilde{c}}{\lambda_0} \text{ (permitivity } \mu = 1), \tag{9}$$

Equation (8) is convenient because it permits the l integration to be done trivially, yielding

$$(\mathrm{R_{abs}})_f \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E_{\text{ in}}|^2 \cdot \pi R^2 \cdot 2 \int_0^1 dC C \left(1 - e^{-2n_i k_o l} f\right), \tag{10a}$$

$$(\mathrm{R_{abs}})_b \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E_{\text{ in}}|^2 \cdot \pi R^2 \cdot 2 \int_o^1 dC C e^{-2n_i k_o l} f \left(1 - e^{-2n_i k_0 (l_b + l_b')}\right), \tag{10b}$$

Here $\langle T \rangle$ is an average transmission factor, and the dimensionless ratio

$$C \equiv c/R, \tag{11}$$

$(\mathrm{R_{abs}})_f$ = rate of energy absorption in front hemisphere, and similarly for $(\mathrm{R_{abs}})_b$. The subscripts f and b stand for the front and back hemisphere, respectively. Note that in each case the absorption rate can be written as

$$\mathrm{R_{abs}} \simeq \begin{pmatrix}\text{transmission}\\ \text{factor}\end{pmatrix} \begin{pmatrix}\text{incident}\\ \text{flux}\end{pmatrix} \begin{pmatrix}\text{cross} - \text{sectional area}\\ \text{of absorber}\end{pmatrix} \begin{pmatrix}\text{attenuation}\\ \text{factor}\end{pmatrix}, \tag{12}$$

To proceed further, we note that simple geometric analyses yield

$$\begin{aligned} l_b &= RF \frac{\left\{C^2 + \left[\sqrt{1 - C^2} + F\right]^2\right\}^{1/2}}{F + \sqrt{1 - C^2}} \\ l_f &= l_b \cdot \frac{\sqrt{1 - C^2}}{F} \\ l_b' &= 2R \left[\frac{1 + 2BF + B^2F^2}{1 + 2BF + F^2}\right]^{\frac{1}{2}}, \end{aligned} \tag{13}$$

where

$$F \equiv \frac{f}{R}, \quad B \equiv \sqrt{1 - C^2}. \tag{14}$$

Combining equations (10)–(14), and using a simple polynomal fit, we obtain

$$\begin{aligned} (\mathrm{R_{abs}})_f \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E^{\text{ in}}|^2 \pi R^2 &\left\{1 - \frac{2}{3} \exp\left[-\frac{\sqrt{1 + F + F^2}}{1 + 2F} x\right]\right\} \\ &- \frac{1}{3} \exp\{[-x]\}, \end{aligned}$$

$$
\begin{aligned}
(\mathrm{R_{abs}})_b &\simeq \langle T\rangle \frac{\tilde{c}}{8\pi}|E^{\text{ in }}|^2\pi R^2 \cdot 2\left\{\frac{1}{3}\exp\left[-\frac{\sqrt{1+F+F^2}}{1+2F}x\right]\right\} \\
&\quad + \left\{\frac{1}{6}\exp[-x] - \beta_0\left(\frac{1}{2}+\frac{1}{3}\beta_1+\frac{1}{4}\beta_2\right)\right\}, \\
\beta_0 &\equiv e^{-2x/\sqrt{1+F^2}}, \\
\beta_1 &\equiv -3 + \frac{4}{\beta_0}\left(\exp\left[-\frac{(2+F)}{\sqrt{1+F+F^2}}x\right] - \frac{1}{4}\exp\left[-2x\right]\right), \\
\beta_2 &\equiv \frac{1}{\beta_0}\exp\left[-2x\right] - \beta_1 - 1, \\
x &\equiv 2n_i k_o R. \qquad (15)
\end{aligned}
$$

It is interesting that in our approximation the absorption rates depend on n_r, n_i, and k_o only through F and $x = 2n_i k_o R$. In particular, once F if specified, the rates essentially depend only on the dimensionless quantity $2n_i k_o R$.

Let us define $\hat{R}_f$ and $\hat{R}_b$ by the relation

$$
(\mathrm{R_{abs}})_f \equiv \langle T\rangle \frac{\tilde{c}}{8\pi}|E \text{ in }|^2\pi R^2\ \hat{R}_f, \qquad (16)
$$

and a similar one for $\hat{R}_b$. Then $\hat{R}_f$ and $\hat{R}_b$ can be calculated from equations (15). The results for $n_r = 1.5 (F = 1)$ are shown in Figure 2.

As we shall see in more detail later, we can expect the photophoresis effect to be positive or negative depending on whether more energy is absorbed in the forward or backward hemisphere. Hence Figure 2 indicates that negative photophoresis occurs for small x and positive for large x. This is just what is expected: At small x, focusing dominates attenuation => negative photophoresis, and vice versa at large x.
According to Figure 2, approximately ~25% more energy can be absorbed in the back hemisphere than in the front hemisphere for $n_r = 1.5$, for the most favorable values $2n_i k_o R \simeq 0.3$.

As a check on the possible accuracy of our analyses we can compare our results with the experimental data quoted by Pluchino (1983). According to Pluchino, experimental data for $n_r = 1.57$ (about the value used above) indicate that the crossover from negative

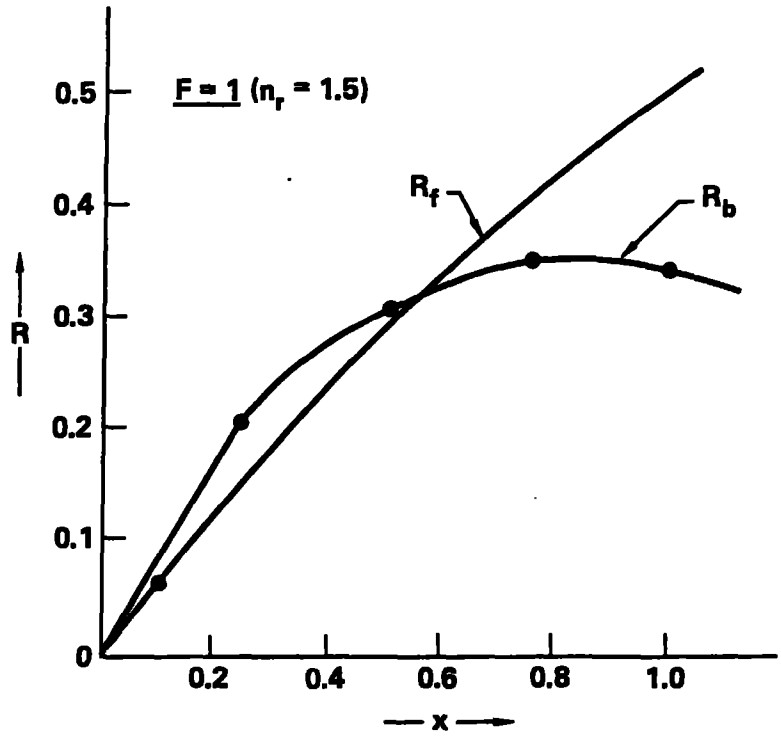

Figure 2. Forward and backward absorption factors.

to positive photophoresis for $n_r = 1.57$ and $n_i = 0.038$ occurs at $k_0R \simeq 11$. Hence $x = 2n_ik_0R \simeq 0.84$. Our predicted crossover value, from Figure 2, is $x \sim 0.6$ or so. Hence the agreement is not bad. Later we shall discuss further the agreement of the predictions of our analyses with Pluchino's data.

For soot particles we also need expressions for R_{abs} when $F > 1$ and especially when $n_r - 1 << 1$ and $n_i << 1 (Eq.\ 3)$. So consider a situation with $F > 1$. Leaving out the details, analyses parallel to the above yield

$$(R_{abs})_f \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E_{\text{ in}}|^2 \pi R^2 \cdot 2 \int_0^1 dCC \left[1 - e^{-2n_ik_ol_f}\right], \tag{17}$$

$$(\mathrm{R}_{\mathrm{abs}})_b \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E_{\text{ in}}|^2 \pi R^2 \cdot 2 \int_0^1 CCe^{-2n_ik_ol_f} \left(1 - e^{-2n_ik_o\tilde{l}_b}\right),$$

where $l_f = \dfrac{L\sqrt{1-C^2}}{F + \sqrt{1-C^2}} R,$

$$\tilde{l}_b = 2\left[1 - \frac{C^2F^2}{L^2}\right]^{\frac{1}{2}} R - \frac{L\sqrt{1-C^2}}{F + \sqrt{1-C^2}},$$

$$L = \left[(F + \sqrt{1 - C^2})^2 + C^2\right]^{1/2}. \tag{18}$$

For soot particles we are interested in situations with $F >> 1$ and $x << 1$. Hence, combining equations (17) and (18), and expanding in x and $\frac{1}{F}$, we obtain

$$(\mathrm{R_{abs}})_f \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E_{\text{ in}}|^2 \pi R^2 \cdot \left[\frac{2}{3}x(1 + \frac{1}{5F^2}) - \frac{1}{4}x^2(1 + \frac{1}{3F^2})\right]$$

$$(R_{abs})_b \simeq \langle T \rangle \frac{\tilde{c}}{8\pi} |E_{\text{ in}}|^2 \pi R^2 \cdot \left[\frac{2}{3}x(1 + \frac{3}{4F} - \frac{1}{5F^2})\right]$$

$$-\frac{3}{4}x^2 \left[\left(1 + \frac{32}{45F} + \frac{1}{3F^2})\right)\right]. \tag{19}$$

In order to apply these results we need expressions for the photophoretic force. We turn to this problem next.

B. The Net Photophoretic Force

According to the calculations of Yalamov et al. (1976) the net photophoretic force F_{ph} acting on a soot particle irradiated by the sun is given by

$$\frac{F_{ph}}{F_{grav}} \simeq \frac{1.1 \times 10^4 \tilde{T}}{e^{-.129h} R_p^{0.2}} \frac{(\tilde{T}/288)}{(\chi/0.02)} \tag{20}$$

Here F_{grav} is the gravitational force acting on the particle, $\tilde{T}$ is the temperature in K, χ is the thermal conductivity in MKS units, h is height in km above the earth, and R_p is the particle radius in μm. Further, the crucial quantity J is given by

$$J \equiv \frac{3}{2} n_r n_i k_o R \int \frac{r^3 dr}{R^4} \int d\tilde{\theta} sin2\tilde{\theta} |\frac{E}{E_{\text{ in}}}|^2 \tag{21}$$

Here $\tilde{\theta}$ is the usual spherical polar angle with the z axis taken as the direction of propagation of the incident wave.

In our rough approximation, we will relate J to $(R_{abs})_b - (R_{abs})_f$ by saying

$$J \simeq \frac{3}{2} n_r n_i k_o R |\langle \frac{r}{R} \frac{sin2\tilde{\theta}}{sin\tilde{\theta}} \rangle| \int (d^3 r |\frac{E}{E_{\text{ in}}}|^2 sign(sin2\tilde{\theta}), \tag{22}$$

where the brackets denote an average value, taken to be the same for both hemispheres. It follows from Eqs. (8), (9), and (22) that

$$J \simeq \frac{3}{8}\langle T\rangle|\langle\frac{r}{R}\frac{sin2\tilde{\theta}}{\sin\tilde{\theta}}\rangle|(R_b - R_f), \tag{23}$$

Before applying this formula in Eq. (20), let us ask whether it is a reasonable approximation. We do so by comparing its predictions with the experimental data in Figure 5 of Pluchino (1983). The data quoted by Pluchino for $n_r = 1.57$ and $n_i = 0.38$ show that for large $x = 2n_i k_o R$, the factor J tends to a value sin -0.3. But from Figure 2 and Eq. (23) we would say that at large x in this case,

$$\begin{aligned} J &\sim \frac{3}{8}\langle T\rangle|\langle\frac{r}{R}\frac{sin2\tilde{\theta}}{sin\tilde{\theta}}\rangle|(0-1) \\ &\sim -0.38\langle T\rangle|\langle\frac{r}{R}\frac{sin2\tilde{\theta}}{sin\theta}\rangle|. \end{aligned} \tag{24}$$

Hence again we have fairly good rough agreement.

Returning to Eqs. (19), (20), and (23) for soot particles, and employing Eq. (3) for the index of refraction of 0.1 μm soot particles, we have

$$\frac{F_{ph}}{F_{grav}} \simeq 20\frac{e^{.129h}}{(R/0.1\mu m)}k_o R\{1 - 0.7(k_o R))\} < T > \frac{\times|\langle\frac{r}{R}\frac{sin2\tilde{\theta}}{sin\tilde{\theta}}\rangle|(\tilde{T}/288)}{(\chi/0.02).}$$

We can obtain an upper limit by setting the two averages to unity. Since the terminal velocity achieved is proportional to the force, we see that the photophoretic velocity is greater than the gravitational velocity by factors ranging up to $\sim 10^2$ and that for $koR \leq 1$, the photophoresis is negative. Thus the largest negative photophoretic velocities expected for small soot particles are

$$\begin{aligned} (V_{ph})_{max} &\sim 20 \times 2 \times 10^{-4} \text{ km/week} \\ &\sim 40 \times 10^{-4}\text{km/week}. \end{aligned} \tag{25}$$

For large soot particles the imaginary part of the index of refraction is smaller, so the photophoretic effect is smaller. Hence we conclude that the photophoretic effect is unimportant for soot.

3. Light Scattering and the Designed Aerosol Problem

This problem involves the question concerning what types of material, and in what particle shapes, absorb and scatter radiation at various wavelengths most efficiently.

A large group of workers have used a variety of numerical methods to decipher the extinction properties of nonspherical dielectric bodies at various wavelengths. I have made some progress on the application of a "Born series" technique to this problem. At the center of this approach is the fact that a formal solution for the displacement $D = \epsilon E$ in a dielectric body is

$$D = D_0 + \frac{\epsilon - \epsilon_o}{\epsilon} \nabla \times \nabla \times \int_{\text{body}} \frac{e^{ik_o|x-x'|}}{4\pi|x-x'|} D' d^3x', \tag{26}$$

Here D^o is the incident plane field and k_o its wave number, ϵ = dielectric constant of body, and ϵ_0 = dielectric constant of surrounding medium.

I have discovered (for myself at least) that if the integral is expanded in power of $k_o l$, where l = size of body, then precisely for generally nonaxisymmetric ellipsoids, Eq. (26) can be solved, in principal analytically, order by order in kl; and, further, that at the nth order the expression for D involves polynomials (in the coordinates) of order n and nothing higher.

I have succeeded in solving through zero and first order, and I have an approximate solution at 2nd order—the set of algebraic linear equations one ends up having to solve is becoming quite large.

The important point, I think, is that solving through 2nd order will give one the first two terms, those proportional to $k_o l$ and $(k_o l)^3$, for the absorption cross section σ_{abs}, for artitrary dieletric constant ϵ. This should enable one to get a good estimate of σ_{abs} all the way up to the absorption peak, as the behavior is quite smooth in all known numerical examples. This approximate behavior can be matched to the known larger kl behavior, which says that the cross-section per unit volume falls off like $(k_o l)^{-1}$, at large kl. This

should yield the approximate location of the absorption peak as well as a fair fit across the whole spectrum.

I am confident that the results will support the conclusion that aluminum-like thin disks are the best infrared absorbers.

What about scattering? Here needle-like objects can have larger cross sections. But it is not clear whether scattering processes can do the trick as far as strong influence on a nuclear-winter scenario is concerned. This is partly because in scattering by nonspherical particles, the differential cross section is diminished in the backward direction (i.e. toward earth for radiation emitted from earth); and, also, the radiation may eventually escape after multiple scattering. Hence careful analysis of the transfer problem is called for.

Acknowledgments

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