Modelling photoluminescence from interstellar dust

Giuliano Malloci[1,2], Giacomo Mulas[1] and Francesca Useli[2]

[1] INAF–Osservatorio Astronomico di Cagliari, Astrochemistry Group, Strada n. 54, Loc. Poggio dei Pini,
I–09012 Capoterra (CA), Italy
[2] Dipartimento di Fisica, Università degli Studi di Cagliari, Strada Provinciale Monserrato–Sestu
Km 0.700, I–09042 Monserrato (Ca), Italy

E-mail: `gmalloci@ca.astro.it,gmulas@ca.astro.it,fuseli@ca.astro.it`

Abstract. We recently developed a general recipe to extrapolate the expected photoluminescence (PL) of small particles from the knowledge of available laboratory results obtained on bulk samples. Starting from the semi-classical formalism of inelastic scattering by single molecules embedded in small particles, we model the emission as a distribution of uncorrelated oscillating electric dipoles, whose density is a functional of the locally absorbed energy. The PL spectrum is found to depend on the size of the dust particle and on the angle between the incoming exciting light and the direction of observation. We expect the present model to be a useful tool which will allow the study of PL phenomena of interstellar dust to move beyond the straight and possibly misleading comparison with experimental data obtained for bulk samples. We discuss the astrophysical implications of our results, their impact on current hypotheses on the carriers of the Extended Red Emission and their relevance in testing dust rotational properties.

1. Introduction

Interstellar dust is a diluted dispersion of grains of sizes comparable to, or smaller than, the wavelength of visible light, probably with a complex structure (core-mantle, fluffy etc.) (see e. g. [1]). Regardless of their actual shape, such small particles have optical properties which are very different from those of the bulk material they are made of. Therefore, any attempts to quantitatively compare photoluminescence (PL) signals from interstellar particles with experimental data *must* carefully take into account the effect of particle size and optical properties: the difference between the PL properties of bulk samples and those of a small particle may be about as large as that between the respective extinction properties, just for the same reasons. In particular, PL will undergo self-absorption and scattering *within* the material before leaving it, and this effect will be just as different between a dust grain and a bulk sample as absorption and scattering are. In [2] we presented a mesoscopic theory of the PL by small dust grains which enables one to extrapolate laboratory results obtained for bulk samples and obtain the expected PL from small particles made of the same material, very much in the same way as absorption and scattering properties of small dust grains are usually computed using bulk optical "constants" [3]. The detailed treatment of self-absorption renders the PL spectrum dependent both on the size of the dust particle and on the angle between exciting light and the direction of observation. In [4] we discuss the simple implementation of the above model for homogeneous spheres with specific reference to the so-called Extended Red Emission (ERE), a general PL phenomenon originating from interstellar dust particles (see e. g. [1]). In [2, 4] we applied the model only to a specific hydrogenated amorphous carbon (HAC) flavour obtained as an organic refractory residue produced by UV irradiation of an ice mixture [5]. Here we apply the same approach to silicon carbide (SiC) particles, using the optical properties given for α–SiC by [6]. Section 2 recalls from [2, 4] the physics and equations relevant for the present application. In Sect. 3 we discuss the astrophysical implications of our results, their impact on current hypotheses on the carriers of the Extended Red Emission and their further relevance in testing the rotational properties of interstellar dust particles. Finally, Sect. 4 draws the main conclusions of the present work.
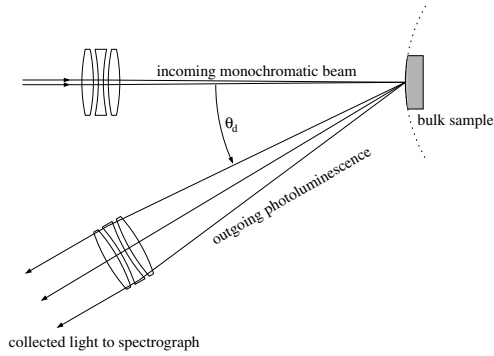
Figure 1. Schematic description of the experimental setup to measure the PL yield $\frac{d\eta_{lab}}{d\Omega d\omega'}$ on a bulk sample and hence obtain $p_{\text{loc}}(\omega, \omega')$ in the local approximation. The sample can be seen as a portion of a sphere of very large radius, represented by the dotted outline.

2. The model

We want to evaluate the impact of particle size effects in modulating the spectral distribution of the PL observed in laboratory experiments performed for bulk samples. We start from the semi–classical theory of inelastic scattering by single molecules embedded in small particles [7]. The elementary PL emission of a volume element dV of the dust particle is represented as an incoherent, unpolarised density of oscillating electric dipoles. This density is assumed to be given by a function of the energy *locally* absorbed from the exciting radiation field in the same volume element dV. Under this *local case* approximation [2, 4], the PL power emitted by a dust grain per unit solid angle and unit frequency interval upon excitation by a monochromatic radiation flux $I(\omega)$, can be written as

$$\frac{dP_{\text{pl}}}{d\Omega d\omega'} = I(\omega) \left(\frac{dC_{\text{pl}}}{d\Omega d\omega'} \right)_{\text{dust}}, \tag{1}$$

where $\frac{dC_{\text{pl}}}{d\Omega d\omega'}$ is the differential cross section for PL computed for the assumed geometry of the particle. This quantity expresses the angular distribution of the light emitted by the particle: the amount of light (per unit incident irradiance) emitted at frequency ω' by the particle into a unit solid angle about a given direction. The quantitative ralation between the experimentally measured PL yield on macroscopic samples and the PL expected from dust particles made of the same material can be expressed as:

$$\frac{dC_{\text{pl}}}{d\Omega d\omega'} = \frac{d\eta_{\text{lab}}}{d\Omega d\omega'} \cdot A(\omega, \omega', a, \theta, \theta_d). \tag{2}$$

The experimentally measured PL yield $\frac{d\eta_{\text{lab}}}{d\Omega d\omega'}$ is the ratio between the measured PL power per unit solid angle and unit frequency in a given direction, as shown in the experimental configuration depicted in Fig. 1, and the power absorbed by the sample from the monochromatic incident exciting light beam. The form factor $A(\omega, \omega', a, \theta, \theta_d)$, which has the dimensions of an area, contains all the small particle effects, including their angular dependence and their dependence on the complex refractive index of the material. The dependence of $A(\omega, \omega', a, \theta, \theta_d)$ on the specific laboratory setup used to measure the PL yield is explicitly represented here by the angle θ_d. For the particular case of a homogeneous sphere [2], the angular dependence of $A(\omega, \omega', a, \theta, \theta_d)$ is given in terms of an expansion in Legendre polynomials:

$$A(\omega, \omega', a, \theta, \theta_d) = \frac{1}{g(\omega, \omega')} \sum_t \gamma_t(a, \omega, \omega') P_t(\cos \theta), \tag{3}$$

whose coefficients γ_t contain the dependence on the sphere radius a and the refractive index N_1 at ω and ω'. The factor $g(\omega, \omega')$ in the above formula depends on the experimental setup used to measure the PL and takes into account self-absorption and scattering within the laboratory sample.
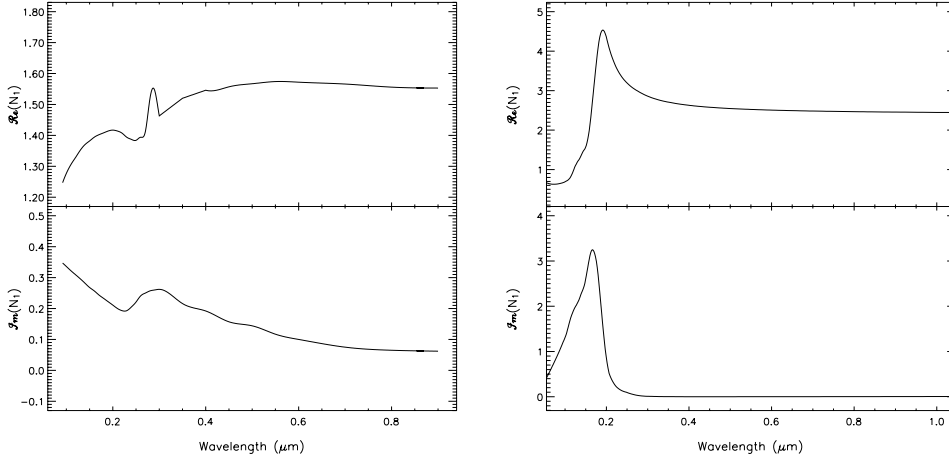
Figure 2. Real and imaginary part of the refractive index of HAC from [5] (left panel), and α-SiC from [6] (right panel).

As astrophysically relevant examples we consider the optical constants given for HAC by [5] and those of α-SiC taken from [6] (see Fig. 2). In the case of carbonaceous materials the excitation and subsequent PL are known to be effectively confined to a single graphitic platelet (see e. g. [8]), providing a firm justification for using the local approximation in [2, 4]; we use the same approximation to describe the PL arising in irradiated SiC, which is clearly due to localised defects, although confinement of the excitation still needs to be established. Fig. 3 displays the behaviour of $A(\omega, \omega', a, \theta, \theta_d)$ at fixed sphere radius assuming monochromatic excitation $\lambda = 2\pi c/\omega$ at 220 nm for HAC and 266 nm for α-SiC. $A(\omega, \omega', a, \theta, \theta_d)$ shows a marked dependence both on the wavelength of the emitted radiation λ' and on the angle θ between the direction of the incident light and the direction of observation of the PL. The first dependence is clearly a consequence of the behaviour of the assumed refractive indices, which causes increasing self-absorption at smaller wavelengths (see Fig. 2). The second dependence can be seen, in a naïve intuitive way, as an indication of the varying path length that the outgoing light must travel in the absorbing medium before escaping the particle. Both of these dependences display an interference pattern superimposed on a smoother overall behaviour. The interference pattern is strongly dependent both on λ and a hence it will tend to average out and disappear if one considers either a broad distribution of particle sizes or of exciting wavelengths, or both. In the case of a distibution $n(a)$ Eq. (1) becomes:

$$\frac{dP_{\text{pl}}}{d\Omega d\omega'} = \int da\, n(a) I(\omega) \left(\frac{dC_{\text{pl}}}{d\Omega d\omega'} \right)_{\text{dust}}. \tag{4}$$

The resulting effective $\bar{A}(\omega, \omega', \theta, \theta_d)$ obtained assuming a distribution of particle sizes $n(a) \propto a^{-3.5}$, with 10nm $\leq a \leq$ 1000 nm [9], are shown in Fig. 4. As expected, the interference pattern seen for fixed sizes in Fig. 3 is washed out; in addition, $\bar{A}(\omega, \omega', \theta, \theta_d)$ displays a marked dependence on the angle θ.

3. Astrophysical implications

Interstellar particles are non-spherical, as evidenced by the observed polarisation of starlight; moreover, they are not homogeneous, since they cyclically experience very different ambient conditions, being subjected to the deposition (inside dense clouds) of frozen mantles, energetic processing by UV radiation and cosmic rays, sublimation of volatile components (in the diffuse medium), and grain-grain collisions leading to accretion and shattering. Nonetheless, since a distribution of spherical, homogeneous dust particles *does* successfully reproduce the main observational characteristics of the extinction curve, we do expect our simple model to exhibit, to the same extent, the main qualitative effects of dust grain geometry and self-extinction on their PL properties. More detailed and realistic models will be needed for a more precise quantitative evaluation, but still the present one is a first step in the right direction.
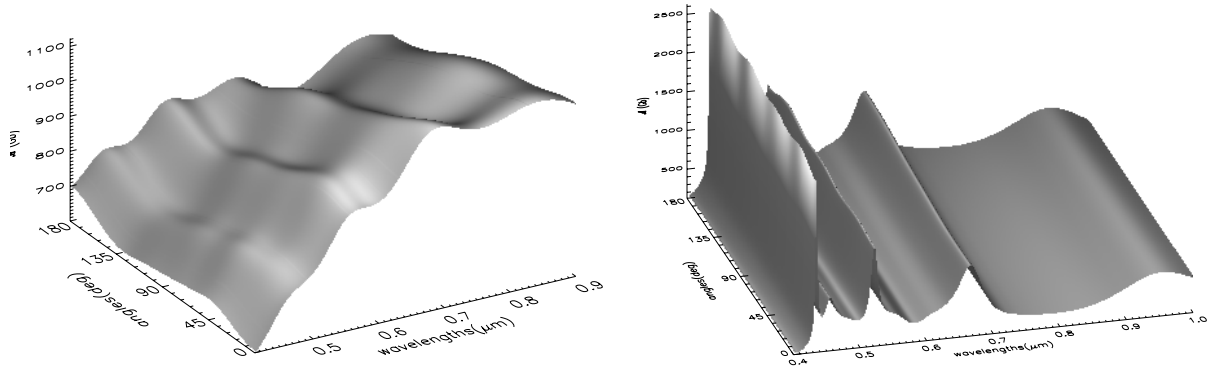
Figure 3. Form factor $A(\lambda, \lambda', a, \theta, \theta_d)$ obtained for $a = 350$ nm, $\lambda = 220$ nm and the optical constants of HAC [5] (left panel) and $a = 180$ nm, $\lambda = 266$ nm and the optical constants of α-SiC of [6] (right panel).

The astrophysical implications of our simplified model are related to the explicit angular dependence of the PL spectrum obtained. One basic question relevant from the astrophysical point of view is: "how much does a dust particle rotate in the time interval it spends between the absorption and the subsequent emission processes?". If the above rotation $\Delta\theta$ is large, the dust particle will lose all memory of the direction the exciting photon came from; in this case, the outgoing PL will be modulated by *the angular average* of $A(\omega, \omega', a, \theta, \theta_d)$. If, on the other hand, this rotation is small, the particle can be considered to be essentially "frozen" between absorption and emission, and the outgoing PL will be modulated by $A(\omega, \omega', a, \theta, \theta_d)$, including its full angular dependence. In intermediate cases, one should consider a weighted angular average of $A(\omega, \omega', a, \theta, \theta_d)$ over the distribution of rotation angles of the particle between absorption and emission. The order of magnitude of the average $\Delta\bar{\theta}$ can be estimated as:

$$\Delta\bar{\theta} = \bar{\omega}_{\mathrm{rot}}\,\tau, \tag{5}$$

where τ is the typical decay time of an electronic transition involving emission of radiation, and $\bar{\omega}_{\mathrm{rot}}$ is the characteristic angular frequency of rotation of such a dust particle in interstellar conditions. In turn, we may estimate $\bar{\omega}_{\mathrm{rot}}$ considering the particle as a rigid rotating body with momentum of inertia I in thermal equilibrium with the kinetic temperature of the gas T_{gas}:

$$\frac{1}{2}I\,\bar{\omega}_{\mathrm{rot}}^2 = k_B\,T_{\mathrm{gas}}, \tag{6}$$

where k_B is the Boltzmann constant. The momentum of inertia of a homogeneous sphere in terms of its radius a and its density ρ_0 is $8\pi\rho_0/15\,a^5$, which, combined with Eqs. (5) and (6), gives $\Delta\bar{\theta}$ in radians as:

$$\Delta\bar{\theta} = \bar{\omega}_{\mathrm{rot}}\tau = \left(\frac{15}{4\pi\rho_0}\right)^{1/2}\left(\frac{k_BT_{\mathrm{gas}}}{a^5}\right)^{1/2}\tau \sim 128.4\left(\frac{g/cm^3}{\rho_0}\right)^{1/2}\left(\frac{T_{\mathrm{gas}}}{K}\right)^{1/2}\left(\frac{\mu m}{a}\right)^{5/2}\left(\frac{\tau}{s}\right).$$

Assuming a temperature $T = 100$K for the diffuse interstellar medium (ISM), a particle density of the order of 1 g/cm³, and a decay time of the excited state of the order of 10^{-8} seconds, we get for $\Delta\bar{\theta}$ values of about $2.3 \cdot 10^{-6}$, $7.3 \cdot 10^{-5}$, $2.3 \cdot 10^{-4}$ radians, for a ranging from 2, to 0.5 and 0.05 μm, respectively. In all these cases, the estimated $\Delta\bar{\theta}$ is much smaller than the scale of the detailed angular structure of $A(\omega, \omega', a, \theta, \theta_d)$ hence these particles can be considered to be effectively immobile between absorption and emission. In this case, $A(\omega, \omega', a, \theta, \theta_d)$, with no angular average, will be the function modulating the outgoing PL. The above still holds even for temperatures as high as $T \sim 10^4$K, since $\Delta\bar{\theta} \propto T^{1/2}$, hence for a very wide range of physical conditions where the ERE is observed. For extremely small

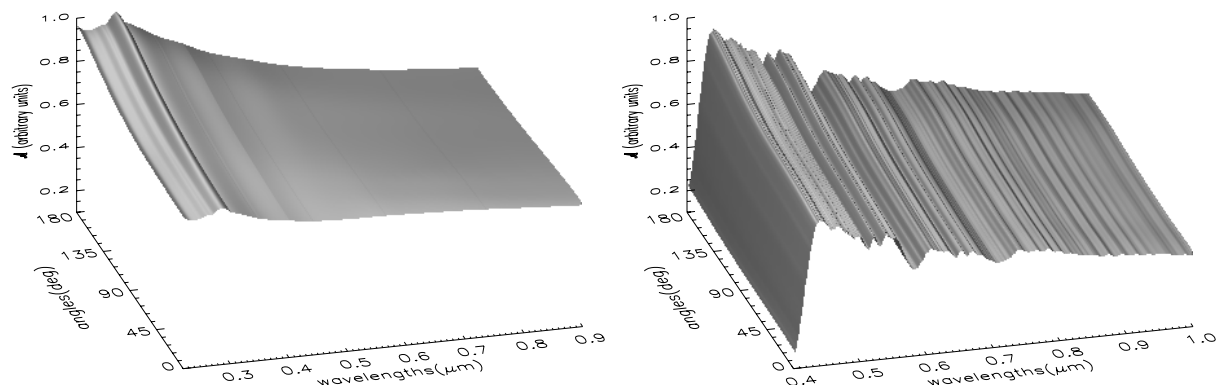**Figure 4.** Weighted average of $A(\lambda, \lambda', a, \theta, \theta_d)$ over a size distribution $n(a) \propto a^{-3.5}$ with 10 nm $\leq a \leq$ 1000 nm for HAC (left panel) and α-SiC (right panel) excited at 220 nm and 266 nm, respectively.

sphere radii, namely 0.005μm = 5nm, we obtain for $\Delta\bar{\theta}$ a value of about 7 radians which *does* produce a strong angular mixing in the emission process such that any memory of the direction of the exciting light is lost. However, such small particles will be in the Rayleigh limit anyway, for any reasonable optical constants; hence, in this specific case, $A(\omega, \omega', a, \theta, \theta_d)$ shows no angular dependence anyway, and dust grain rotation makes no difference as far as the resulting PL is concerned.

Summing up, in a typical interstellar environment spherical particles in the range of sizes between 0.05 and 2μm should not rotate appreciably between the absorption and subsequent emission of a photon, *provided that their rotation is in thermal equilibrium with the kinetic temperature of the gas.* In such a situation a memory of the direction of the incident radiation is conserved in the emission process, and an explicit knowledge of this dependence is fundamental in the application to PL phenomena originating from interstellar dust particles such as ERE. Indeed, one of the properties of the ERE is that its spectral shape is observed to be different even in different positions of the same object [10]. Our work suggests that this effect may be at least partly due to the variation of the angle between the observation direction and the direction the exciting radiation comes from, something that was overlooked in previous analyses. However, at least part of the dust grain particles have long been thought to rotate much faster than thermally: [11] first thoroughly calculated the effects of the so called *rocket effect*, whereby inhomogeneities of a particle lead to torques which do not average to zero over time. A prototypical example of the rocket effect is dust-catalysed H_2 formation [12]. This effect was long thought to dominate suprathermal dust grain rotation, until [13] first provided a thorough calculation of radiative torques for a model anisotropic particle, showing that these latter torques may even dominate those due to H_2 formation in the case of relatively large dust particles and/or strongly anisotropic radiation fields. The current state of the art of theoretical work as to dust grain rotation in the ISM seems to indicate that:

- very small particles may rotate subthermally due to efficient damping via microwave emission [14];

- particles smaller than $\sim 0.1\mu$m will rotate thermally, since neither radiative torques [13] nor catalytic H_2 formation [15] are able to significantly spin them up;

- particles larger than $\sim 0.1\mu$m will be driven by radiative torques to rotational speeds which can exceed by a few orders of magnitude those expected in thermal equilibrium conditions ("suprathermal" rotation) [13].

In principle, therefore, for dust particles larger than $\sim 0.1\mu$m rotational speeds might be large enough to wash out at least some of the angular dependence of $A(\omega, \omega', a, \theta, \theta_d)$. While such suprathermal rotation of interstellar dust grains has a strong theoretical foundation and is believed to be important in several astrophysical conditions, there is no direct observational evidence to date of its existence. Our model

predicts a direct consequence of suprathermal rotation of dust grains in astronomical PL phenomena such as ERE, which would be amenable to direct testing.

4. Conclusions

This overly simplified application of our model, even with its acknowledged limitations, still provides an important improvement over neglecting particle size dependent self-absorption effects altogether, as almost all studies comparing the ERE with laboratory data on the PL of carbonaceous materials did so far. While is not yet possible to draw definitive conclusions from the present indicative results, nonetheless some rather strong points can be safely made:

- self-absorption and geometric effects *must* be taken into account when comparing laboratory PL data taken on macroscopic samples with the observed PL originating from small interstellar particles;

- we suggest that the major objections against the hypothesis of HAC as the carrier of the ERE, namely that it is impossible to simultaneously match both the spectrum and the PL efficiency, must be reexamined: either appropriate laboratory measures should be made *directly* on small particles similar in size and structure to the ones believed to be present in astrophysical environments, or a theoretical model like the one we presented should be used to bridge the gap between the macroscopic laboratory samples and the interstellar dust grains. Indeed, our work hints that small particle effects would make the PL arising from dust grains considerably redder, maybe enough to bring the PL spectrum arising from highly hydrogenated HAC into acceptable agreement with ERE observations, while still preserving their high quantum efficiency. For a definitive assessment of this possibility the present model will need to be generalised to more realistic dust particles;

- carbonaceous dust particles, which are included essentially in all dust models, must be strongly photoluminescent in environments where they are hydrogenated, and thus their expected emission must be at least *compatible* with ERE, or the models will need to be revised accordingly.

- SiC grains are found in meteorites and were identified as a component of interstellar and circumstellar dust from the IR feature at about 11.3 μm; since laboratory experiments show solid SiC to be strongly luminescent, its emission must be shown to be compatible with observations.

References

[1] Draine B T 2003 *ARA&A* **41** 241-289
[2] Malloci G, Mulas G and Benvenuti P 2004 *A&A* **420** 809-820
[3] Borghese F, Denti P and Saija R 2003 *Scattering from model nonspherical particles* (Berlin: Springer-Verlag)
[4] Mulas G, Malloci G and Benvenuti P 2004 *A&A* **420** 921-927
[5] Jenniskens P 1993 *A&A* **274** 653-661
[6] Laor A and Draine B T 1993 *ApJ* **402** 441-468
[7] Chew H, McNulty P J and Kerker M 1976 *Phys. Rev. A*, **13** 396-404
[8] Seahra S S and Duley W W 1999 *ApJ* **529** 719-723
[9] Mathis J S, Rumpl W and Nordsieck K H 1977 *ApJ* **217** 425-433
[10] Witt A N and Boroson T A 1990 *ApJ* **355** 182-189
[11] Purcell E M 1979 *ApJ* **231** 404-416
[12] Rouan D, Léger A and Le Coupanec P 1997 *A&A* **324** 661-673
[13] Draine B T and Weingartner J C 1996 *ApJ* **470** 551-565
[14] Draine B T and Lazarian A 1998 *ApJ* **508** 157-179
[15] Lazarian A and Draine B T 1999 *ApJ* **516** L37-L40