COLLECTIVE EXCITATIONS AND

DUST PARTICLES IN SPACE

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ABSTRACT

Due to collective processes small solid particles exhibit very strong resonances in absorption and scattering in the spectral regions in which ε_1, the real part of the dielectric constant, is negative. The precise positions depend on the particle shape, size and on the nature and amount of coatings it might have. A comprehensive discussion of these resonances is given in the Rayleigh approximation. It is shown that the observed bands at 2200 Å and in the 10 μ region are most probably due to such collective excitations. Some other processes are also considered and are shown to be relatively inefficient and inadequate. The following specific conclusions are drawn: (a) the 2200 Å interstellar band is very likely due to graphite particles; (b) these graphite particles should be very small, approximately spherical, and should have no coating whatsoever; (c) the identification of circumstellar and interstellar silicates from the observations in the 10 μ region does not seem to be correct; (d) very valuable information about the shape of the circumstellar and interstellar dust particles can be obtained directly from observations; (e) narrow band polarization measurements in the spectral regions of these bands will be very helpful in determining the shape of the particles.

I.   INTRODUCTION

Recent OAO-2 observations of the interstellar extinction curve have been discussed by Bless and Savage (1972; see also this volume). The observations show large intrinsic varia-

tions. Bless and Savage have concluded that a multi-component
model of interstellar grains, such as I had proposed earlier
(Gilra 1970, 1971), is needed to explain the observations.

The most significant feature in the entire extinction curve
is the ultraviolet bump at about $\lambda^{-1} = 4.6 \ \mu^{-1}$. The observa-
tions indicate that, in general, this feature is very strong,
has a quite symmetrical profile, and is not "flat-topped".
Also, the variations in the position of the extinction maximum
are very small. In this paper our main concern is to identify
the physical process and the substance which can give rise to
such a feature at this wavelength (about 2170 Å).

It seems, as will be shown below, that the most plausible
process which can satisfactorily explain this band is that of
plasma oscillations. This process was introduced in astronomy
by Unsöld (1964) to explain the diffuse interstellar band at
4430 Å. He also mentioned that the position of the resonance
due to this process (plasma resonance) shifts to longer wave-
lengths as the size of the particles is increased, but he did
not explain this effect. It was pointed out by van de Hulst
(1964) that the strength and position of plasma resonances de-
pend on the shape of the particles. Apart from the work of
Unsöld (1964) and that of van de Hulst (1964) there is no dis-
cussion of this process in the astronomical literature. Simi-
lar resonances occurring in the infrared for some solids will
be discussed. Observations also indicate a band in the 10 μ
region (Woolf and Ney 1969, Hackwell et al. 1970).

It will be shown that the theoretical resonances occur due
to "collective" processes. A comprehensive discussion of
these resonances will be given in the small-size limit. In
this limit they can be studied with the help of solutions of
relatively simple problems in electrostatics. The following
cases will be considered: homogeneous ellipsoids, coated el-
lipsoids, and ellipsoids with anisotropic optical properties.
A very brief discussion of the effect of particle-size on the
position of the resonances will be given. The physical pro-
cesses which give rise to these resonances will be mentioned.

Next, the astronomical observations in the ultraviolet and
infrared will be compared with the theoretical results. Some
other physical processes will also be considered and the sug-
gested identifications will be critically examined. Finally,
some observationally verifiable predictions will be made.

II. THEORY

a) Small Homogeneous Ellipsoidal Particles

Let us discuss absorption and scattering by homogeneous el-
lipsoidal particles in the Rayleigh approximation. For a field
applied along one of the main axes (j = 1,2,3) of the ellipsoid,

the polarizability α_j is a tensor and is written as (van de Hulst 1957, 1964)

$$\alpha_j = \frac{V}{4\pi} \frac{\varepsilon - \varepsilon_m}{L_j(\varepsilon - \varepsilon_m) + \varepsilon_m} \quad , \tag{1a}$$

where V is the volume of the particle, ε its dielectric constant, ε_m is the dielectric constant of the medium, the L_j depend on the ratios of the axes, and the values $4\pi L_j$ are known as the depolarization factors. The L_j satisfy the condition $0 \leq L_j \leq 1$, with the property $\sum_j L_j = 1$. If $m = n - ik$ is the complex refractive index, we have $\varepsilon \equiv m^2 = \varepsilon_1 - i\varepsilon_2 = n^2 - k^2 - i2nk$. If the particle is in free space, $\varepsilon_m = 1$, and equation (1a) reduces to

$$\alpha_j = \frac{V}{4\pi} \frac{\varepsilon - 1}{L_j(\varepsilon - 1) + 1} \tag{1b}$$

Equations (1a) and (1b) apply to the case of long elliptical cylinders also (van de Hulst 1957, p. 71).

The scattering cross-section, C^j_{sca}, and the absorption cross-section, C^j_{abs}, are given by

$$C^j_{sca} = \frac{8\pi}{3} \left(\frac{2\pi}{\lambda}\right)^4 |\alpha_j|^2 \quad , \tag{2}$$

and

$$C^j_{abs} = 4\pi \left(\frac{2\pi}{\lambda}\right) \text{Re}(i\alpha_j) \quad , \tag{3}$$

where Re means "the real part of". Putting the value of α_j from (1a) in (2) and (3), we get

$$C^j_{sca} = \frac{8\pi^3}{3} \frac{V^2}{\lambda^4} \frac{(\varepsilon_1 - \varepsilon_m)^2 + \varepsilon_2^2}{\{L_j\varepsilon_1 + \varepsilon_m(1 - L_j)\}^2 + (L_j\varepsilon_2)^2} \quad , \tag{4}$$

and

$$C^j_{abs} = \frac{2\pi}{\lambda} V \frac{\varepsilon_m\varepsilon_2}{\{L_j\varepsilon_1 + \varepsilon_m(1 - L_j)\}^2 + (L_j\varepsilon_2)^2} \quad . \tag{5}$$

Here we have assumed that ε_m is real; we shall also assume that its value does not change with wavelength.

From equations (4) and (5) we see that there is a resonance in the absorption and scattering cross-sections at the wave-

lengths where

$$L_j \varepsilon_1 + \varepsilon_m (1-L_j) = 0 \quad , \tag{6}$$

and $L_j \varepsilon_2 < 1$. This is because, as pointed out by van de Hulst (1964), the polarizability of the particles is very large as can be seen from equation (1a). As we see from equation (6) there is one resonance corresponding to each value of L_j. We therefore have three resonances for an ellipsoid, two for a spheroid and one for a sphere. These resonances occur where ε_1, the real part of the dielectric constant, is negative and are confined roughly within the maximum of ε_2, corresponding to $L_j = 0$, and the maximum of $(\varepsilon_1^2 + \varepsilon_2^2)^{-1}$, corresponding to $L_j = 1$. This is the case of a thin disc. The location of the particle in a medium of dielectric constant ε_m or in free space ($\varepsilon_m = 1$) does not affect the position of these boundaries. However, for intermediate values of L_j, the resonances in the case of free space are at

$$\varepsilon_1 = -\{(1/L_j) - 1\} \quad , \tag{7a}$$

and in the case of a medium at

$$\varepsilon_1 = -\varepsilon_m \{(1/L_j) - 1\} \quad . \tag{7b}$$

When discussing real solids we shall see that these conditions imply a shift in the position of the resonances to lower energies (or longer wavelengths) as the particles are placed in a medium of dielectric constant ε_m because of the way ε_1 varies with wavelength. As an example we take the case of a spherical particle. $L_1 = L_2 = L_3 = 1/3$, and in free space the resonance is at $\varepsilon_1 = -2$, whereas if the particle is in a medium of dielectric constant ε_m, the resonance occurs at $\varepsilon_1 = -2\varepsilon_m$, that is, at a higher negative value of ε_1.

We may remark in passing that C_{sca} has a minimum which occurs on the higher energy side of the resonance. The conditions from equation (4) are $\varepsilon_1 = \varepsilon_m$ and $\varepsilon_2 = 0$. These are the same conditions which give a minimum in the reflectivity, and the frequency at which these are satisfied in the infrared is known as the Christiansen frequency.

b) Size Dependence

As the size of spherical particles is increased the resonance shifts to longer wavelengths (Unsöld 1964). Similar results are obtained for spherical graphite particles (see, e.g., Gilra 1971). For very small sized graphite particles the extinction maximum is at about 2100 Å and as the mean size is increased the feature becomes broader and the maximum shifts

to longer wavelengths. We have to use the Mie theory to explain this (see, e.g., Doyle and Agarwal 1965, Ruppin and Englman 1968). A detailed discussion of size dependence will be given elsewhere (Gilra 1972). We just note that as the first departures from the Rayleigh approximation become apparent and we consider only the electric dipole term, the resonance occurs at a higher negative value of ε_1 given by

$$\varepsilon_1 = -2 - \frac{12}{5} x^2 \quad , \tag{8}$$

where $x = 2\pi a/\lambda$ and a is the radius. This result can be readily obtained from an equation given by van de Hulst (1964, p. 28), though he does not seem to have noticed this.

As the mean size becomes quite large the resonance becomes broad and shallow as can be seen in the extinction curves of SiC (Gilra 1971, Figure 2). The broad shallow feature shortward of about 1800 Å seen in that figure is due to such a resonance. Once again this resonance is to the high energy side of the maximum of ε_2.

c) Effect of Coatings

Let us consider an ellipsoid of dielectric constant ε_i having a coating in the shape of a confocal ellipsoid and a dielectric constant ε_o. Let this compound ellipsoid be placed in free space. Following Stepin (1965) and Bilboul (1969) we can derive an expression for the polarizability, α, for such an ellipsoid. For a field applied along one of the main axes the polarizability is given by

$$\alpha = \frac{V}{4\pi} \frac{(\varepsilon_o-1)\{L_i(\varepsilon_i-\varepsilon_o) + \varepsilon_o\} + \frac{V_i}{V}(\varepsilon_i-\varepsilon_o)\{L(1-\varepsilon_o)+\varepsilon_o)\}}{\{(\varepsilon_i-\varepsilon_o)L_i+\varepsilon_o\}\{1+L(\varepsilon_o-1)\} + \frac{V_i}{V} L(1-L)(\varepsilon_i-\varepsilon_o)(\varepsilon_o-1)}, \tag{9}$$

where V_i is the volume of the inner ellipsoid, V is the total volume, and $4\pi L_i$ and $4\pi L$ are the depolarization factors of the inner ellipsoid and the total ellipsoid respectively. For the sake of clarity we have suppressed the direction dependence. If $V_i/V = 0$, or $V_i/V = 1$, or $\varepsilon_i = \varepsilon_o$, or $\varepsilon_o = 1$, this equation reduces to equation (1b) for the case of homogeneous ellipsoids. If $\varepsilon_i = 1$, we get an expression for the polarizability of a hollow shell whose inner and outer surfaces are confocal ellipsoids.

In the degenerate cases, thin discs, spheres and thin needles, we have $L_i = L$. For coated spheres $L_i = L = 1/3$ and the result is identical to that obtained for the small size case from the rigorous solution (Güttler, 1952, van de Hulst 1957, p. 74). For a thin disc, or a highly flattened oblate spheroid, $L_i = L$ and there are two values of L, L = 0 and

L = 1. The results are quite interesting.
   For L = 0,

$$\alpha = \frac{V}{4\pi} \{ (1-\frac{V_i}{V}) (\varepsilon_o - 1) + \frac{V_i}{V}(\varepsilon_i - 1) \}$$

or, $$\alpha = \{ \frac{V_o}{4\pi} (\varepsilon_o - 1) + \frac{V_i}{4\pi}(\varepsilon_i - 1) \} \quad , \tag{10}$$

and for L = 1,

$$\alpha = \frac{V}{4\pi} \{ (1-\frac{V_i}{V}) \frac{\varepsilon_o - 1}{\varepsilon_o} + \frac{V_i}{V} \frac{\varepsilon_i - 1}{\varepsilon_i} \}$$

or, $$\alpha = \{ \frac{V_o}{4\pi} \frac{\varepsilon_o - 1}{\varepsilon_o} + \frac{V_i}{4\pi} \frac{\varepsilon_i - 1}{\varepsilon_i} \} \quad , \tag{11}$$

where V_o is the volume of the shell and V_i is the volume of the inner ellipsoid. On comparing equations (10) and (11) with equation (1b) we note that the polarizability of such a compound disc (or, for limiting values of L) is equal to the sum of the polarizability of two separate thin discs having the volumes V_o and V_i, and with dielectric constants ε_o and ε_i respectively.

   Scattering and absorption cross-sections in the Rayleigh approximation for compound ellipsoids can be calculated by using the value of polarizability from equation (9) in equations (2) and (3) respectively. The case of a compound thin disc is simple; we can use equations (10) and (11) in equations (2) and (3). For such a thin disc as in the case of polarizability, the absorption cross-section also is the sum of the absorption cross-sections of two separate thin discs having the volumes V_o and V_i, and dielectric constants ε_o and ε_i respectively. For other cases the final expression for C_{abs} and C_{sca} are somewhat complicated if both ε_i and ε_o are complex but they become relatively simple if ε_o is real. Before we write the expressions, let us compare the results for the case of a thin needle, or a highly elongated spheroid (L = L_i; L = 0 and L = 1/2), with those for a coated infinite circular cylinder in the small radius limit.

   From the rigorous solution of Kerker and Matijević (1961) for perpendicular incidence, I have obtained the following dominant terms (up to x^2) in the approximation that the total radius is much smaller than the wavelength both outside and inside the cylinder. For the TE mode (E ⊥ symmetry axis) the

dominant term is

$$a_1 = \frac{i\pi x^2}{4} \left[\frac{(\epsilon_0-1)(\epsilon_i+\epsilon_0) + q^2(\epsilon_i-\epsilon_0)(1+\epsilon_0)}{(\epsilon_i+\epsilon_0)(1+\epsilon_0) + q^2(\epsilon_i-\epsilon_0)(\epsilon_0-1)} \right]$$

$$\equiv \frac{i\pi x^2}{4} A \text{ , say,} \tag{12a}$$

and for the TM mode (E ‖ symmetry axis) it is

$$b_0 = \frac{i\pi x^2}{4} \{(1-q^2)(\epsilon_0-1) + q^2(\epsilon_i-1)\} \text{ ,}$$

$$\equiv \frac{i\pi x^2}{4} B \text{, say,} \tag{12b}$$

where $x = 2\pi r/\lambda$, r is the total radius, $q = r_i/r$, and r_i is the inner radius, and as before, ϵ_0 and ϵ_i are dielectric constants of the shell and core respectively. These reduce in the appropriate limits to the case of a thin homogeneous infinite cylinder. C_{ext}, the extinction cross-section per unit length and C_{sca}, the scattering cross-section per unit length for the two cases are given by

TE mode: $C_{ext} = 2r \cdot \frac{2}{x} \text{Re}(2a_1)$,

$$C_{sca} = 2r \cdot \frac{2}{x} \cdot 2|a_1|^2 \text{ ;} \tag{13}$$

TM mode: $C_{ext} = 2r \cdot \frac{2}{x} \text{Re}(b_0)$,

$$C_{sca} = 2r \cdot \frac{2}{x} |b_0|^2 \text{ .} \tag{14}$$

Let us now compare the cross-sections for a thin infinite circular cylinder with those of a small highly elongated prolate spheroid. The TE mode corresponds to L = 1/2 and the TM mode to L = 0 for the highly elongated prolate spheroid. In terms of A and B, as defined in equations (12a) and (12b), the polarizabilities for a small highly elongated prolate spheroid are,

for L = 1/2, $\alpha = (V/4\pi)2A$, (15)

and for L = 0, $\alpha = (V/4\pi)B$, (16)

where we have made use of the fact that in this case $V_i/V = q^2$. If ℓ is the major axis and r the semi-minor axis of the whole spheroid, $V = 2/3 \pi r^2 \ell$. The absorption and scattering cross-sections can now be calculated with the help of equations (3)

and (2) respectively. We note that for absorbing particles in this approximation $C_{ext} = C_{abs}$ and we can compare the two in a straightforward way.

So that a direct comparison is possible the cross-sections for the two cases, thin infinite circular cylinder and small highly elongated prolate spheroid, are presented in a tabular form. We note that for the former case the tabulated cross-sections are <u>per unit length</u>.

Thin Infinite Circular Cylinder	Small Highly Elongated Prolate Spheroid
TE mode: $$C_{ext} = \frac{2\pi}{\lambda} \cdot \pi r^2 \cdot 2 \cdot [\text{Re}(iA)]$$ $$C_{sca} = \frac{\pi^2 r}{2} \cdot (2\pi r/\lambda)^3 \cdot [|A|^2]$$	L = 1/2: $$C_{abs} = \frac{2\pi}{\lambda} \cdot \frac{2}{3}\pi r^2 \ell \cdot 2 \cdot [\text{Re}(iA)]$$ $$C_{sca} = \frac{8}{27}\pi \ell^2 \cdot (2\pi r/\lambda)^4 \cdot [|A|^2]$$
TM mode: $$C_{ext} = \frac{2\pi}{\lambda} \cdot \pi r^2 \cdot [\text{Re}(iB)]$$ $$C_{sca} = \frac{\pi^2 r}{4} \cdot (2\pi r/\lambda)^3 \cdot [|B|^2]$$	L = 0: $$C_{abs} = \frac{2\pi}{\lambda} \cdot \frac{2}{3}\pi r^2 \ell \cdot [\text{Re}(iB)]$$ $$C_{sca} = \frac{2}{27}\pi \ell^2 \cdot (2\pi r/\lambda)^4 \cdot [|B|^2]$$

C_{ext} and C_{abs} for the two cases are the same; the only difference is that in the corresponding volumes. For the scattering cross-sections the terms in the square brackets, and these are the more important terms, are the same because of the depolarization factor, but the other terms are different. For the infinite cylinder case we have a λ^{-3} dependence, whereas for the highly elongated prolate spheroid the wavelength dependence is λ^{-4}. (For all ellipsoids in general in this approximation the scattering cross-section has a λ^{-4} dependence.) The same is true for the corresponding homogeneous cases (cf., van de Hulst 1957, p. 316; Kerker 1969, p. 266).

We now write expressions for C_{abs} and C_{sca} for the compound ellipsoid on using equation (9) in equations (3) and (2) if ε_o is real.

$$C_{abs} = \frac{2\pi}{\lambda} V_i \frac{\varepsilon_o^2 \varepsilon_2}{(\varepsilon_1 T + \varepsilon_o U)^2 + (\varepsilon_2 T)^2} \quad , \qquad (17)$$

and

$$C_{sca} = \frac{8\pi^3}{3} \frac{V^2}{\lambda^4} \frac{(\varepsilon_1 R + \varepsilon_o S)^2 + (\varepsilon_2 R)^2}{(\varepsilon_1 T + \varepsilon_o U)^2 \qquad (\varepsilon_2 T)^2} \quad , \qquad (18)$$

where we have substituted $(\varepsilon_1 - i\varepsilon_2)$ for ε_i, and

$$R = L_i (\varepsilon_o - 1) + \frac{V_i}{V} \{\varepsilon_o - L(\varepsilon_o - 1)\} ,$$

$$S = (1 - L_i)(\varepsilon_o - 1) - \frac{V_i}{V} \{\varepsilon_o - L(\varepsilon_o - 1)\} = (\varepsilon_o - 1) - R ,$$

$$T = L_i \{1 + L(\varepsilon_o - 1)\} + (\varepsilon_o - 1)\frac{V_i}{V} L(1 - L) ,$$

and $U = (1 - L_i)\{1 + L(\varepsilon_o - 1)\} - (\varepsilon_o - 1)\frac{V_i}{V} L(1 - L) = \{1 + L(\varepsilon_o - 1)\} - T.$

We shall now make the further assumption that ε_o is independent of wavelength. Let us investigate the nature of the resonances. For the limiting cases $L = L_i$ and, $L = 0$ and $L = 1$ and the results can be easily obtained from equations (10) and (11). For C_{abs} the coatings do not have any effect on the position or the strength of the resonances but for C_{sca} the shell is also contributing and there is a slight change. For intermediate values of L, $0 < L < 1$, the resonances occur at a value of ε_1 given by

$$\varepsilon_1 = -\varepsilon_o \frac{U}{T} = -\varepsilon_o \left(-1 + \frac{L + L(\varepsilon_o - 1)}{L_i + L(\varepsilon_o - 1)\{L_i + \frac{V_i}{V}(1 - L)\}} \right). \qquad (19)$$

It follows from equation (19) that if there is no coating the resonance occurs at $\varepsilon_1 = -(-1 + 1/L_i)$ as in the case of a homogeneous ellipsoid. It can be shown that as the relative amount of coating is increased (keeping V fixed and decreasing V_i) the resonance occurs at higher negative values of ε_1. Finally, if $V_i/V \to 0$, the resonance occurs at $\varepsilon_1 = -\varepsilon_o(-1 + 1/L_i)$, corresponding to the case of a homogeneous ellipsoid in a medium of dielectric constant ε_o. Thus whatever the amount of coating the resonance is always between $\varepsilon_1 = -(-1 + 1/L_i)$ and $\varepsilon_1 = -\varepsilon_o(-1 + 1/L_i)$. This result is similar to that for surface plasmons in the case of coated thin foils (Raether 1967) and is more general. (These results apply to the case of a coated thin infinite cylinder also.) As noted earlier, the boundaries of the resonances do not change because of coatings. We also note that the minimum in C_{sca} does not shift from $\varepsilon_1 = 1$ to $\varepsilon_1 = \varepsilon_m$ because the shell is also contributing to the scattering; the shift is actually to the lower energy side.

The result we shall be making use of in this paper is that even a slight coating makes a substantial change in the posi-

tion of the resonance.

Although the formula for polarizability, equation (9), is implicit in the work of Stepin (1965) and Bilboul (1969), most of the discussion in this section is believed to be new.

d) Small Ellipsoids with Anisotropic Optical Properties

Since graphite is a suggested interstellar particle and since its optical properties are highly anisotropic (that is, ε is a tensor), let us discuss the nature of these resonances for such particles in the small size limit. The polarizabilities can be calculated from the work of Jones (1945; see also van de Hulst 1957, p. 73). We consider an oblate spheroid whose axes coincide with the principal directions of the dielectric constant tensor. (The results can be extended to the more general cases.) If $\varepsilon_\perp$ and $\varepsilon_{||}$ are the dielectric constants when $\vec{E}$, the electric vector of the incident radiation, is perpendicular and parallel to the symmetry axis respectively, and $4\pi L_\perp$ and $4\pi L_{||}$ are the respective depolarization factors, the corresponding polarizabilities, $\alpha_\perp$ and $\alpha_{||}$, are given by (Wickramasinghe 1967, Greenberg 1968)

$$\alpha_\perp = \frac{V}{4\pi} \frac{\varepsilon_\perp - 1}{L_\perp(\varepsilon_\perp - 1) + 1}$$

$$\alpha_{||} = \frac{V}{4\pi} \frac{\varepsilon_{||} - 1}{L_{||}(\varepsilon_{||} - 1) + 1}$$

(20)

where V is the volume of the particle. These equations are similar to equation (1b). As before, we can calculate the scattering and absorption cross-sections from equations (2) and (3) for the two cases separately.

Let us write $\varepsilon_\perp = \varepsilon_{\perp,1} - i\varepsilon_{\perp,2}$ and $\varepsilon_{||} = \varepsilon_{||,1} - i\varepsilon_{||,2}$. As in the case of a homogeneous oblate spheroid, there are two resonances, one for $\vec{E}$ perpendicular to the symmetry axis ($\vec{E}\perp c$) and the other for $\vec{E}$ parallel to the symmetry axis ($\vec{E}||c$), assuming, of course, that both $\varepsilon_{\perp,1}$ and $\varepsilon_{||,1}$ become negative in some energy ranges. There are several interesting aspects. Even for a spherical particle, which is one extreme case of an oblate spheroid, there are two resonances, one for $\vec{E}\perp c$ at $\varepsilon_{\perp,1} = -2$, and the other for $\vec{E}||c$ at $\varepsilon_{||,1} = -2$. As the particle becomes oblate the resonance for $\vec{E}\perp c$ shifts to higher negative values of $\varepsilon_{\perp,1}$ and for $\vec{E}||c$ to lower negative values of $\varepsilon_{||,1}$. Finally, for the limiting case of a thin disc (platelet or highly flattened oblate spheroid), the resonances are at the maximum of $\varepsilon_{\perp,2}$ and the maximum of $(\varepsilon_{||,1}^2 + \varepsilon_{||,2})^{-1}$ respectively.

e) Physical Processes

So far we have discussed the nature of these resonances in terms of ε, the dielectric constant. We noted that in spite of the possible changes in the shape, amount of coating, size and optical anisotropy, the resonances are confined within the maximum of ε_2 and the maximum of $(\varepsilon_1^2 + \varepsilon_2^2)^{-1}$. If all these parameters are specified the resonance occurs at a fixed negative value of ε_1, and hence for a given solid, at a fixed energy or wavelength. In other words, the excitations are quantized. The question now arises: what are the energy ranges in which ε_1 is negative for the solids of astrophysical interest?

Depending on the type of solid, ε_1 becomes negative, in general, in two energy ranges (Kittel 1966): one in the infrared due to lattice vibrations and the other in the ultraviolet. The behavior can be quite complicated and in each of these energy ranges there can be more than one such region. In the infrared the frequency corresponding to the maximum of ε_2 is called ω_T, the transverse optical phonon frequency, and the frequency at which $\varepsilon_1 \to 0$ and $\varepsilon_2 \to 0$ is called ω_L, the longitudinal optical phonon frequency. ω_T is less than ω_L. In the ultraviolet the frequency at which $\varepsilon_1 \to 0$ and $\varepsilon_2 \to 0$ is called ω_P, the plasma frequency. There is no frequency corresponding to ω_T, but there is a maximum in ε_2 due to the transitions of bound electrons on the lower energy side of ω_P.

We now very briefly discuss the physical processes involved in the excitation of the resonances we have been discussing in this paper. If infrared photons are incident on a diatomic crystal of ions $\pm e$, the positive and negative ions are displaced with opposite signs. Quantized vibrations that follow are designated as "material polaritons" (Englman and Ruppin 1968a,b; Ruppin and Englman 1968). In the ultraviolet the electrons take the place of negative ions. The electron gas starts moving as a whole with respect to the positive ion background. This is known as plasma oscillation and the quantum of this excitation is termed a plasmon (Kittel 1966). Both these processes can be characterized as collective excitations. Thus the resonances discussed earlier in this paper can be identified as occurring due to collective processes. Two qualifications should be made, however. The peak in ε_2 in the ultraviolet is, as noted earlier, due to the transitions of bound electrons. Therefore the resonances occurring at this energy for some shapes are not due to a collective process. In the case of gold and silver in the visual and near ultraviolet, for example, there is what is known as the free carrier plasma frequency, but there is no corresponding peak in ε_2 on the lower energy side of it. The discussion of the resonances has to be modified somewhat to take this fact into

consideration for such cases.

The discussion of the physical processes involved has been, of necessity, very simplified. For details reference should be made to papers cited in this section (see also, Kawabata and Kubo 1966, Steinmann 1968).

Let us now summarize all the theoretical discussion on these resonances. They are due to collective processes, with the exceptions noted above, and are confined within ω_T and ω_L in the infrared and within the maximum of ε_2 and ω_p in the ultraviolet. The precise position depends on the shape, size and amount of coating. We can invert this argument. From an observed feature, if it can be established that these processes are involved, we can determine, at least in principle, the shape and the size of the particles and also how much coating these particles have.

III. COMPARISON WITH OBSERVATIONS

a) The 2200 Å Band

As earlier calculations indicated there is an extinction band for small-sized spherical graphite particles at about 2200 Å. This has been attributed to transition of π electrons to the conduction band in the graphite crystal (Wickramasinghe 1967, Stecher 1969). This identification is incorrect because this band occurs where $\varepsilon_{\perp,1}$ is -2, and is therefore due to plasma oscillations in spherical graphite particles. The dielectric constant of graphite in the basal plane, $\varepsilon_\perp$, is given by Taft and Phillip (1965, and private communication) and is plotted in Figure 1 for energies between about 2.5 ev and 9.0 ev. The plasma frequency, ω_p, is at about 1800 Å (7.0 ev). The maximum in ε_2 is at about 2800 Å. For spherical particles the resonance is at $\varepsilon_1 = -2$ which is at 2205 Å. For an oblate spheroidal particle with the ratio of minor to major axis = 0.8 the resonance shifts to 2255 Å. Similarly, if we coat a spherical graphite particle of 50 Å radius with a shell of ice about 5 Å thick, the resonance shifts to 2255 Å.

In Figure 2 is plotted the absorption cross-sections using equations (20) for graphite particles for two extreme shapes, a sphere and a thin disc, normalized such that they are unity at the maximum. If $C_{abs}^\perp$ and $C_{abs}^{\parallel}$ are the absorption cross-sections on using $\varepsilon_\perp$ and $\varepsilon_\parallel$ respectively, the quantity plotted is $(2C_{abs}^\perp + C_{abs}^{\parallel})/3$, as is the case if the particles are randomly oriented. Values of $\varepsilon_\parallel$ for graphite were taken from Tosatti and Bassani (1970). There are no strong resonances in $C_{abs}^{\parallel}$.

The highest negative value of $\varepsilon_1^{\parallel}$ is -0.85 which occurs at 12.7 ev. In this approximation $C_{abs}^{\parallel}$ is greater than $C_{abs}^\perp$ between about 1300 and 850 Å, the exact wavelengths depending on
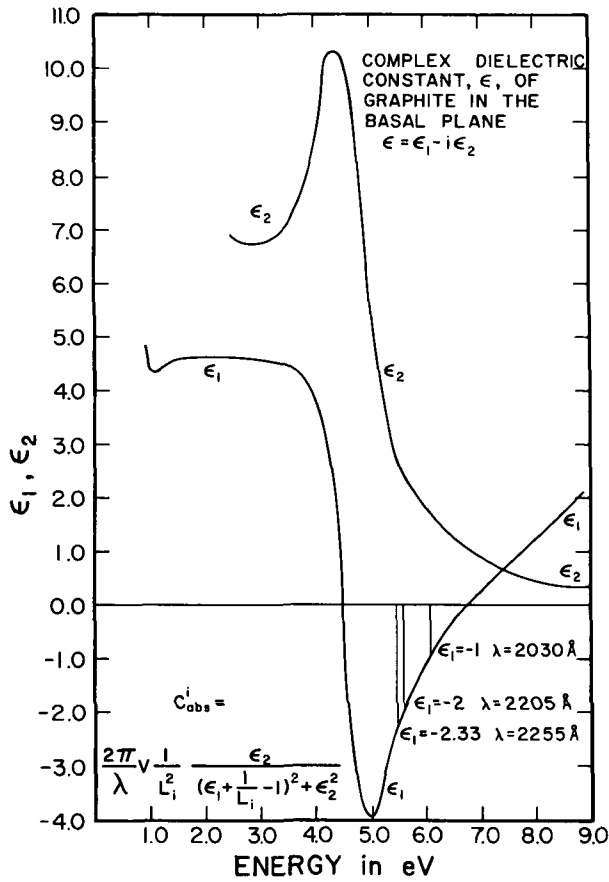
Figure 1. — The complex dielectric constant of graphite in the basal plane (see text for discussion).

the shape. The net effect is to increase somewhat the total C_{abs} in this wavelength range from what is obtained from $C_{abs}^{\perp}$. But at these wavelengths the Rayleigh approximation is not valid and we cannot say what the contribution is for larger sizes because the required theory is not available. It may be remarked that in $C_{abs}^{\perp}$ there is a much stronger resonance in the far ultraviolet than the one at 2070 Å. This resonance occurs at about 800 Å and is also due to the process of plasma oscillations.

The peak in the small size limit as calculated with equations (20) occurs at 2080 Å for spheres; however, $\varepsilon_{\perp,\perp}$ is equal to -2 at 2205 Å. The shift to shorter wavelengths could occur for two reasons. From the formulas we see that the resonance occurs where ε_2 also satisfies the condition, $\varepsilon_2 < 1$. It occurs at shorter wavelengths if this condition is not

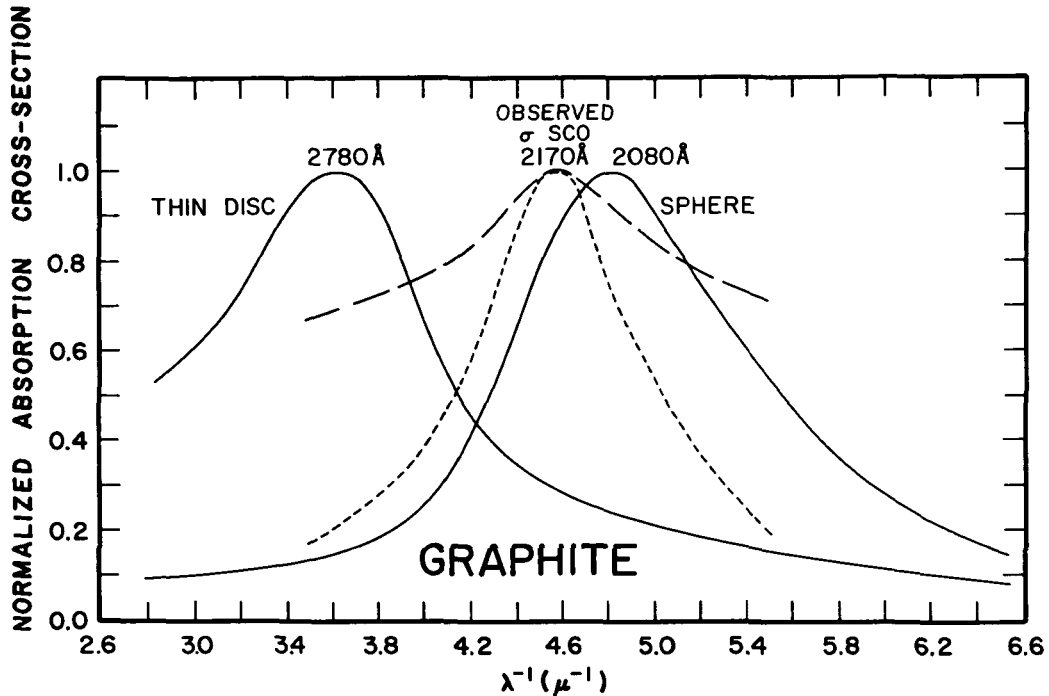Figure 2.—Shape dependence of resonances in graphite. For comparison the observed extinction curve for σ Sco is also plotted using two different base lines (see text for discussion).

satisfied (see, Kawabata and Kubo 1966). The second factor is that C''_{abs} is also contributing and it is increasing towards shorter wavelengths, the net effect being to push the resonance to shorter wavelengths. The second factor is not important in this case, however.

Two other computational details should be noted about such resonances in general. If one does not anticipate where the resonances will be and does not compute cross-sections for a sufficient number of wavelengths, one is likely to get misleading results. Secondly, one should use the dielectric constant and not the refractive index to get physical and computational insight about these resonances.

The peak for spherical particles occurs at 2080 Å and as the oblateness increases it shifts to longer wavelengths. Finally, for thin discs it occurs at 2780 Å. Also plotted in Figure 2 is the observed interstellar extinction curve in the region 2860-1840 Å for σ Sco (Bless and Savage 1972) using two different base lines for its normalization. The broader one (large dashes) is obtained if the base line is parallel to the abscissa and starts at the furthest point observed in the

infrared. The narrower one (small dashes) is obtained if the base line is a smooth curve joining 2.6 μ^{-1} and 6.5 μ^{-1} in the extinction curve of σ Sco. The width of the observed feature depends critically on the choice of the base line. The peak in the observed curve is at 2170 Å and the feature is quite narrow.

It is obvious from Figure 2 that the observations put very stringent limits on the shape of the particles. We can definitely rule out graphite platelets (thin discs) as being responsible for the observed feature.

My calculations for spherical particles, using the Mie theory with $\varepsilon_\perp$, show that for a radius of 0.005 μ the peak is at 2070 Å, for 0.010 μ at 2100 Å, and for 0.020 μ at 2200 Å. If we now compare the theoretical curves taking all the factors into consideration with the observed curves, we can conclude that the suggested interstellar graphite particles should be fairly spherical, quite small (mean radius $\approx$ 0.015 μ) and have almost no coatings whatsoever. The minimum in the observed curves between about 1800-1500 Å is also consistent with the graphite hypothesis (Gilra 1971).

If we take the values of ε from Scouler (1969) for Mg_2Si, the calculations show a strong feature at about 2200 Å for spherical Mg_2Si particles due to plasma oscillations. The list could be made substantially bigger but it is not clear if the particles of other type will be formed anywhere.

As we have noted, the position and strength of the resonance depend critically on the precise values of $\varepsilon(\lambda)$. However, in the discussion so far we have tacitly assumed that we can use the bulk dielectric constants for particles in interstellar space. This may not be exactly true: for sizes $\lesssim$ 100 Å, ε depends on size due to quantum effects which tend to broaden the plasma resonance (Kawabata and Kubo 1966); we do not know what effect the temperature (the temperature of interstellar particles is ~20°K) has on ε; the interstellar particles may not be identical to bulk crystals in their crystalline structure. Surface "irregularities" also affect the nature of plasma resonances (Steinmann 1968). We still do not know how to calculate the extinction cross-section for particles with anisotropic optical properties if the size is arbitrary. The effects of these and perhaps some other related problems should be investigated.

Another suggestion to explain the 2200 Å band is that it is caused by an absorption edge. Huffman and Staap (1971) experimentally obtained the optical constants for one silicate, $(MgFe)SiO_3$, up to 12.4 ev. They suggested that the observed feature could be explained with a gaussian size distribution having a mean size 0.06 μ. In Figure 3 are plotted extinction (E), scattering (S) and absorption (A) cross-sections for spherical particles of this silicate for two mean radii,
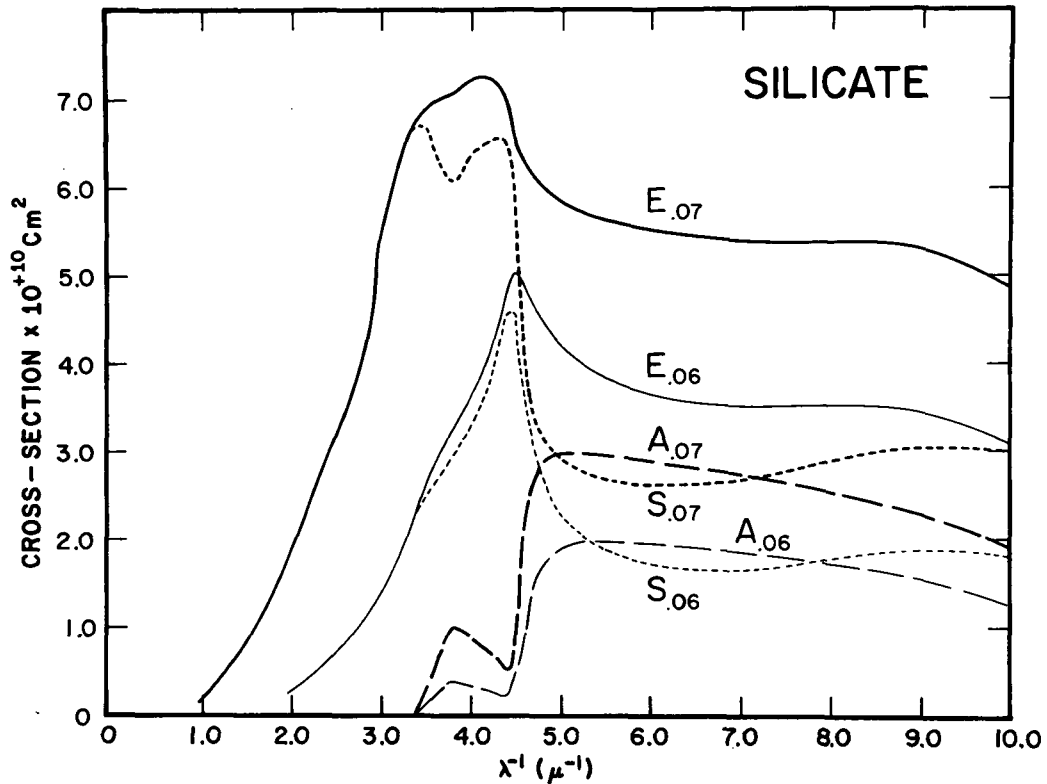
Figure 3.—Theoretical extinction (E), scattering (S) and absorption (A) cross-sections for (MgFe)SiO₃. The suffixes refer to the mean radii in microns (see text for discussion).

0.06 μ and 0.07 μ. The suffixes refer to the mean radii in microns. The curve $E_{.06}$ is the same which according to Huffman and Staap explains the observed feature.

At an absorption edge the imaginary part of the refractive index (or the imaginary part of the dielectric constant) increases rapidly. At this energy the absorption cross-section increases very rapidly and the scattering cross-section decreases very rapidly, as can be seen from the curves. For very small-sized particles the extinction is entirely due to absorption. The relative contribution from scattering increases as the mean size is increased. For a narrow size-range a "band" appears in the scattering cross-section due to the rapid decline on the high energy side (as already noted) and a rapid rise on the low energy side. With only a 15% increase in the mean size, the rapid rise on the low energy side occurs at much lower energies making the scattering "band" extremely broad. The resulting "band" in the extinction cross-

section is "flat-topped", asymmetric and extremely broad and bears no resemblance in shape, strength or position with the observed feature. Therefore it seems unlikely that such a well-defined observed feature can be caused by such a physical "process". Wickramasinghe and Nandy (1971b) have also noted some of these points independently.[*]

Another physical process could be considered. Some solids either due to an intrinsic property or due to an impurity or due to "physical damage" exhibit absorption bands in their bulk optical properties. For example, neutron irradiated quartz shows a band at about 2200 Å (Billington and Crawford, Jr. 1961). Let us investigate if such particles can explain the observed feature. (Dr. G. H. Herbig also mentioned this possibility during the discussion following my paper.) Van de Hulst (1957, p. 191) has already discussed such a theoretical problem. Let us refer to Figure 3. The feature in the absorption cross-section at 3.8 μ^{-1} is due to such a process. Only in the very small-size limit does it show up in the extinction cross-section. As the size increases the <u>minimum</u> in the scattering cross section makes it invisible. In fact, for the mean size 0.07 μ there is a <u>minimum</u> in the extinction cross-section at this wavelength, as already discussed by van de Hulst. It could still be argued, however, that only very small-sized quartz particles may be responsible for the observed feature. However, there is a stronger band in neutron irradiated quartz at 1650 Å which is not seen in the observed extinction curves. Also, since quartz has an absorption edge at somewhat higher energies (the scanner #1 on board the OAO has a quartz window and that is why no observations are possible with it shortward of about 1800 Å), the observed extinction curve should show a very rapid increase at higher energies. The observations of σ Sco and ρ Oph down to 1100 Å (Bless and Savage 1972) which show strong 2200 Å features belie this expectation. Thus it does not seem likely that such particles are responsible for the observed band.

Let us summarize the discussion on the identification of

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[*] One remark should be made about my model of interstellar grains (Gilra 1971). In this model silicates are the chief contributor to the far ultraviolet rise. This conclusion was based on the assumption, which seemed most expedient at the time, that the optical constants are wavelength independent. As discussed here the optical constants are now available for one silicate. The calculations show that the suggested existence of silicates to explain the far ultraviolet rise may be somewhat uncertain now (see Bless and Savage (1972) for other possibilities).

the 2200 Å band. Three different physical processes were considered: plasma oscillations, absorption edges, and absorption bands in bulk crystals. It has been shown that the last two are relatively inefficient and inadequate. In these cases the behavior of scattering is almost opposite to that of absorption in the region of interest (Figure 3). As the size is increased beyond a certain critical value the extinction band becomes exceedingly broad and highly asymmetric in one case, and invisible in the other. On the other hand, the band due to plasma oscillations arises because there is almost identical structure in the bands in both scattering and absorption. Whatever changes we may make in the particle shape (Figure 2), size (Figures 1 and 2 in Gilra 1971) and coatings (as discussed earlier) the extinction band is always well defined and is very strong in the small-size limit. It can be concluded, therefore, that of all the suggestions made so far the process of plasma oscillations is the most plausible process to give rise to a feature of the type observed. It is correct, however, that quite stringent limits were placed on the geometrical and surface properties of the suggested interstellar graphite particles because the observed feature is quite narrow. Some theoretical problems were mentioned earlier and they should be investigated. It may be noted that the minimum in the observed curves between about 1800-1500 Å is also consistent with the graphite hypothesis (Gilra 1971).

b) The Bands in Infrared

It has been suggested (Friedemann 1969a,b; Gilman 1969) that particles of silicon carbide can form in the atmospheres of stars in which the carbon to oxygen ratio is ≥ 1. In the spectra of many of these stars the violet region is strongly depressed, the quasi-discontinuity setting in at about 4400 Å. It has been suggested (Stephenson and Ross 1970) that this phenomenon is "among the most outstanding of all opacity effects to be seen in stars of any spectral type." Silicon carbide has the fundamental absorption edge between 2.2 and 3.0 ev, the precise position depending on the polytype, impurities and temperature. Gilra and Code (1971, 1972) have suggested that circumstellar silicon carbide grains are responsible for the observed violet depression. Their theoretical results are in very good accord with the observations. The model of interstellar grains I have proposed (Gilra 1971) has silicon carbide as a major constituent. We are therefore interested in investigating the nature of resonances in small silicon carbide particles in the infrared so that the observations could confirm or disprove the suggested existence of such circumstellar and interstellar particles.

The fundamental absorption band in SiC is at 12.6 μ

(Spitzer et al. 1959a,b), that is, ω_T, the transverse optical
phonon frequency, corresponds to this wavelength. ω_L, the
longitudinal optical phonon frequency, corresponds to 10.3 μ.
(The optical properties of cubic and hexagonal SiC are quite
similar in this region and we shall consider only the cubic
form.) My Mie calculations for spherical particles had shown
(Gilra 1971) a weak band at ω_T but this was completely dwarfed
by a very strong absorption feature at ~10.75 μ where ε_1 = -2.
I had suggested that the feature will become broad if the par-
ticles are not exactly spherical. At that time I was not
aware that experimental results existed (Pultz and Hertl 1966)
which confirm this prediction. The theory has been discussed
in several papers by Englman and Ruppin (Englman and Ruppin
1968a,b; Ruppin and Englman 1968).

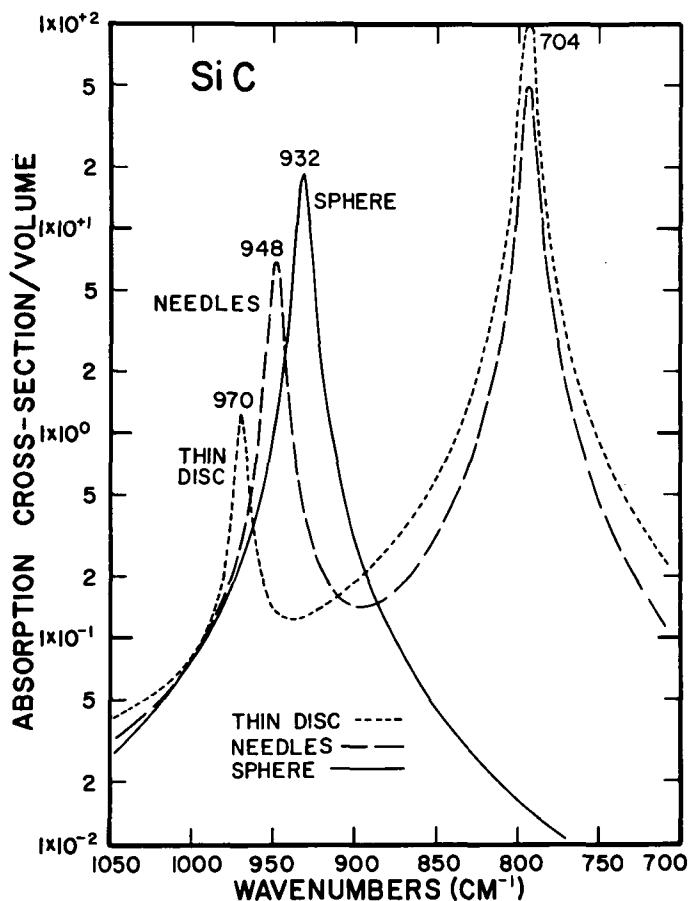Figure 4 shows the absorption cross-section for cubic SiC

Figure 4.—Shape dependence of resonances in SiC in the 10-13 μ
region (see text for discussion). 704 in the upper right
corner should read 794.

particles of three shapes, thin disc, sphere and needles, using equation (5) with $\varepsilon_m = 1$. Random orientation has been assumed. For a thin disc the resonances are at 970 cm^{-1} (= 10.3 μ = ω_L) and at 794 cm^{-1} (= 12.6 μ = ω_T). As the oblateness decreases the resonances move towards each other and finally for the spherical case there is only one resonance at 932 cm^{-1} (= 10.73 μ; $\varepsilon_1 = -2$). As the particle becomes prolate, the resonances move away from each other and finally for the case of a needle they are at 794 cm^{-1} (= 12.6 μ = ω_T) and 948 cm^{-1} (= 10.55 μ; $\varepsilon_1 = -1$). If the particles are "highly irregular" there will be a broad feature between about 10.3 μ and 12.6 μ such as has been observed experimentally by Pultz and Hertl (1966). The results for needles are also in very good agreement with their experimental results for fibres.

We have no a priori way of determining the shape of the suggested circumstellar and interstellar silicon carbide particles. There should be a strong absorption band (or bands) between 10 μ and 13 μ in the interstellar extinction curves of the highly reddened stars. This feature should appear in emission in the stars which show strong violet opacity. However, in the latter case this predicted emission will be somewhat "filled in" by strong absorption in this wavelength region due to SiC$_2$ molecules (which give rise to the Merrill-Sanford bands in the late N-type carbon stars) and high spectroscopic resolution will be needed to separate the two effects (Gilra and Code 1972).

c) The 10 μ Band and Silicates

A broad emission feature centered at about 10 μ has been seen in the spectra of oxygen-rich late type variables (Woolf and Ney 1969). Perhaps a similar feature at 10 μ has been seen in absorption in the observations of the galactic center (Hackwell et al. 1970). These features have been attributed by the authors to circumstellar and interstellar silicates respectively. Since the dielectric constant for silicates is not yet available in this spectral region, let us investigate the theoretical basis for these identifications.

If n, the real part of the refractive index, is very close to one, and k, the imaginary part, is very much smaller than one, then equation (5) reduces to (van de Hulst 1957) with $\varepsilon_m = 1$

$$C_{abs} = (2\pi/\lambda)V \cdot 2k$$

and since the absorption coefficient $A = (4\pi k)/\lambda$, we get

$$C_{abs} = V \cdot A . \qquad (21)$$

Equation (21) gives the absorption cross-section in the Ray-leigh-Gans approximation. This is the equation used by Woolf and Ney (1969) and Hackwell et al. (1970), following the work of Gaustad (1963), in identifying the silicates. Donn et al. (1970) have agreed with this assumption. But this is not correct because the conditions n $\simeq$ 1 and k $<<$ 1 are not satisfied for silicates in this region. The structure in ϵ_1 and ϵ_2 should be somewhat similar to that for quartz (experimental absorptance results (Lyon 1963) show this very well) and that means very high values of ϵ_2 and ϵ_1 (both positive and nega-tive for ϵ_1). The dielectric constant for quartz is given by Spitzer and Kleinman (1961) and theoretical calculations (Knacke 1968, for spherical particles, Martin 1971, for sphe-rical and cylindrical particles, and my own for ellipsoidal particles) using equation (5) show no match at all with the values given by Gaustad (1963) using equation (21). (Most of the values in these spectral regions given by Gaustad (1963) are incorrect for other substances as well, for the same rea-sons.) There are strong resonances (as in the case of SiC) which depend on the shape of the particles. The same should be the case for silicates. For silicates Wickramasinghe and Nandy (1971a) have taken the value of k from Gaustad (1963) and assumed a wavelength independent value of n (= 1.66) throughout this spectral region. This assumption is also not correct. We thus conclude that the identification of circum-stellar and interstellar silicates from observations in the 10 μ region is based on a wrong theoretical premise and inade-quate experimental data and therefore seems to be premature.

There is one way, however, in which the experimental ab-sorption coefficient can be compared directly with the obser-vations, and that is if we assume that the distribution of shapes and sizes of the particles and their spatial arrange-ment are identical in the two cases. It is not clear if this is the case. The best way to resolve the controversy is to get experimental values of ϵ and to obtain observations at high spectroscopic resolution to see the structure within the broad feature. Then the theoretical curves using the values of ϵ obtained experimentally can be directly compared with the observations.

Some of the points in this section have been independently discussed by Martin (1971).

d) Polarization

From equation (5) we see that absorption, and therefore thermal emission, depend on the shape and orientation of par-ticles. If the particles have any anisotropy, either in shape or in optical properties, and if they are suitably aligned, then the 2200 Å band and the bands in the infrared should be

polarized.   There should be a polarity reversal at some wave-
length in these spectral regions.

If the observed interstellar polarization in the visual is
due to the same agent which is responsible for the 2200 Å
band, then this band must be very strongly polarized, as
Stecher and Donn (1965) have already pointed out for graphite.
Recent balloon-borne wide band polarimetry of the reddened
star ζ Oph by the University of Arizona astronomers (Coyne et
al. 1971, and remarks at the 135th meeting of the American
Astronomical Society) indicates that the polarization contin-
ues to decrease in the region 3000-2000 Å.   If these observa-
tions are correct and if the observations of ζ Oph can be con-
sidered as representative of other stars as well, then we can
conclude that the suggested interstellar graphite grains are
not aligned and the polarization in the visual is not due to
graphite grains.

If for the late-type variables the observed polarization in
the visual is due to grains and the emission in the infrared
is also due to the same grains and these grains are suitably
aligned, then the emission feature(s) should be polarized with
a polarity reversal at a fixed wavelength.   However, very nar-
row band polarimetric observations will be required so that
the polarizations with different polarity do not cancel each
other.   Dr. M. Dyck at the Kitt Peak National Observatory has
started such an observational program.

In the model of interstellar grains I have proposed (Gilra
1971) SiC particles are responsible for the observed inter-
stellar polarization in the visual.   As discussed earlier SiC
particles have resonances in the 10-13 μ regions.   Therefore
the suggested interstellar band(s) in the 10-13 μ region should
be polarized with a polarity reversal in between.

It is obvious that narrow band polarization measurements in
these spectral regions will give us important information about
the shape, size and chemical composition of circumstellar and
interstellar grains.

IV.   FINAL REMARKS

Very strong resonances in absorption and scattering occur
in the spectral regions in which ε_1, the real part of the
dielectric constant, is negative.   The precise positions depend
on the particle shape and size and on the nature and amount of
coating.   The nature of these resonances was discussed with
the help of solutions of relatively simple problems in electro-
statics and the discussion given in this paper is essentially
complete in the small-size limit.   In this limit they are al-
ways confined within the maximum of ε_2 on the low energy side
and the maximum of $(\varepsilon_1^2 + \varepsilon_2^2)^{-1}$ on the high energy side.   As
the size becomes comparable to the wavelength, new resonances

appear on the low energy side of the peak in ε_2 (see Ruppin and Englman 1968, Englman and Ruppin 1968b). These will be discussed elsewhere (Gilra 1972).

There are two distinct energy ranges in which these resonances occur. One is in the infrared in the region of strong lattice bands between ω_T and ω_L. The other is in the ultraviolet between the maximum of ε_2, which is due to the transitions of bound electrons, and ω_p. Broadly speaking, the process which gives rise to these resonances is that of collective excitations.

Taking all the factors into consideration and comparing the properties of these resonances with those of "bands" due to other processes, it can be concluded that these resonances are the strongest and, relatively speaking, the most permanent features in the whole domain of light scattering by small particles.

The discussion given in this paper can be applied to some problems in solid state physics, colloidal chemistry and astronomy. With these results it has been possible to explain (Gilra 1972) some experimental results in solid state physics and colloidal chemistry which have not yet been accounted for.

The conclusions concerning some astronomical problems are very interesting. It has been shown that the observed bands at 2200 Å and in the 10 μ region are most probably due to collective excitations. Graphite particles continue to be the best working hypothesis for the 2200 Å band, even though strict limits have been placed on their geometrical and surface properties because of the narrowness of the observed band. A very important result is that for the first time, at least in principle, it may be possible to determine the shape of the circumstellar and interstellar particles directly from observations. Narrow band polarization measurements in the regions of these bands will be very helpful in achieving this goal.

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