QUENCHED CARBONACEOUS COMPOSITE. III. COMPARISON TO THE 3.29 MICRON INTERSTELLAR EMISSION FEATURE

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ABSTRACT

In a previous paper we have shown that oxidized f-QCC (filmy quenched carbonaceous composite) has absorption features that correspond to that of the interstellar 3.3, 3.4, 6.2, 7.7, 8.6, and 11.3 μ m emission features. Here we present new laboratory data showing that oxidized f-QCC, after heating to 500° C, has a 3.29 μm absorption feature that matches precisely the wavelength of the 3.29 μm interstellar emission feature. In addition, the width of the f-QCC feature is close to that of the 3.29 μ m emission feature observed in NGC 7027, Orion, and IRAS 21282+5050. Laboratory spectra of polycyclic aromatic hydrocarbons (PAHs) were also obtained, and comparison of the f-QCC and PAH absorption spectra to that of the 3.29 μm emission feature indicates the f-QCC provides a much better match. We therefore suggest that f-QCC is representative of the class of material giving rise to the emission features in the interstellar medium.

Subject headings: infrared: spectra - interstellar: grains - line identifications

I. INTRODUCTION

Considerable attention has been placed on explaining the origin of the unidentified infrared (UIR) emission features at 3.3, 3.4, 6.2, 7.7, 8.6, and 11.3 μ m that are observed in a variety of astronomical sources (reviewed by Allamandola 1984; Willner 1984; Léger and d'Hendecourt 1987). Additional weaker emission features have been observed by Cohen, Tielens, and Allamandola (1985), Cohen et al. (1986), de Muizon et al. (1986), Nagata et al. (1988), Roche, Aitken, and Smith (1988), and Witteborn et al. (1989), and a listing of most of these features is given by Allamandola, Tielens, and Barker (1987).

The proposed identifications are numerous and include surface functional groups (Duley and Williams 1981), polycyclic aromatic hydrocarbons (PAHs) as summarized by Léger and d'Hendecourt (1987), quenched carbonaceous composite (QCC) (Sakata et al. 1984, 1987), hydrogenated amorphous carbon (Duley and Williams 1988), amorphous carbon (Borghesi, Bussoletti, and Colangeli 1987), carbonaceous residue from meteorites (Wdowiak, Flickinger, and Cronin 1988), mixtures of carbonaceous and PAH materials (Blanco, Bussoletti, and Colangeli 1988), and vitrinite, a form of demineralized coal (Papoular et al. 1989).

Although the PAH identification has received much attention, there have been recent criticisms of this identification by Salisbury et al. (1988), Donn, Allen, and Khanna (1989), Mathis (1988), and Tokunaga et al. (1989) in regard to the lack of agreement with high spectral resolution PAH spectra, with the ultraviolet spectrum of the interstellar medium, and with the spectrum of the diffuse interstellar bands. There are also questions regarding the formation of the infrared continuum by PAHs. In the context of the PAH identification, Léger and d'Hendecourt (1987) and Allamandola, Tielens, and Barker (1985) suggest that a mixture of PAHs gives rise to the UIR emission bands, since the 7.7 and 8.6 μ m bands are much too broad to be produced by any single PAH. However, it has not been demonstrated in the laboratory that a specific mixture of PAH's can produce the UIR emission features, and this has prevented a definitive identification based upon PAHs.

Recent high spectral resolution observations by Nagata et al. (1988) and Tokunaga et al. (1988) have revealed new information on the UIR emission features: the peak wavelength is $3.295 \pm 0.005 \ \mu m$, and the FWHM is $0.043 \pm 0.005 \ \mu m$ for NGC 7027, IRAS 21282 + 5050, and BD $+ 30^{\circ}3639$. In the case of HD 44179 and Elias 1, the peak wavelength is the same (to within measurement errors), but the FWHM is smaller, $0.023 + 0.002 \mu m$. These observations require laboratory data of much higher spectral resolution and signal to noise than previously presented, and this was one of the reasons for the new laboratory studies presented here.

In this paper, we present new laboratory data at 3 μ m that support the idea that the UIR emission features arise from a class of material called QCC and previously discussed by Sakata et al. (1984, 1987). The 3.3 and 11.3 μ m UIR emission features undoubtedly arise from the C-H bond on aromatic molecules, as initially proposed by Duley and Williams (1981) and Léger and Puget (1984); however, QCC is a different type of material than other proposed identifications for the UIR emission features, including PAHs.

In the § II, we present laboratory spectra of QCC that has been heated to 500° C and spectra of various types of PAHs. Our goal is to understand in greater detail how the spectrum of QCC is modified upon heating, and to compare the spectra of the UIR emission features, QCC, and PAHs at approximately the same spectral resolution. The latter is particularly important in testing the validity of the QCC and PAH identifications. Discussion and implications are presented in § III.

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II. LABORATORY STUDIES

a) Thermal Alteration of Oxidized f-QCC

A thin film of oxidized f-QCC was produced in the same manner as described in Sakata et al. (1987). The sample was collected on an NaCl substrate within the apparatus that was used to produce the f-QCC. To study the effect of high temperature on oxidized f-QCC, the sample and NaCl substrate were placed in a specially made fused quartz container. The quartz container was then heated under vacuum in an electric furnace for 30 minutes at a fixed temperature ($\pm 2^{\circ}$ C). After heating, the sample was cooled to room temperature and placed in a JASCO IR-810 infrared spectrometer to measure its spectrum. This instrument has a spectral resolution of 3.5 cm^{-1} . To measure the effect of heating at the next temperature, the sample is placed in the quartz container again and the procedure is repeated. The infrared absorbance at $\sim 25^{\circ}$ C (room temperature) and those after heating to 400°, 450°, and 500° C are shown in Figure 1. The absorbance is the quantity $-\log(I/I_0)$, where I is the transmission of the sample and I_0 is that of the reference, and it is proportional to the absorption coefficient of the sample. The gain of the spectrometer was increased from the room temperature sample to the 500° C sample, as can be seen in the increasing level of the noise. The peak of the 3.29 μ m absorbance peak is ~8 times smaller at 500° C than in the room temperature sample.

At room temperature f-QCC is a brownish film, and the wavelength of the peak absorbance is 3040 cm^{-1} (3.289 μ m). After heating to 250° C the peak absorbance shifts to 3035 cm^{-1} (3.295 μ m). At above 300° C the f-QCC material turns to a dark black color; however, there is no further shift in the peak absorbance. Between 300° and 540° C, the peak absorbance remains at 3035 cm^{-1} . The feature at 11.4μ m, which arises from the "solo" C-H bond on a carbon ring (Sakata *et*



FIG. 1.—Spectra of oxidized f-QCC at room temperature and after heating to various temperatures.

al. 1984), also remains at 540° C. The shift in the 3.3 μ m band from 3.289 to 3.295 μ m indicates a change in the structure of the f-QCC material, perhaps the formation of "cross-linked" structures (see § III*a*). When QCC is heated to above 500° C, the absorbance peak becomes weaker and the material becomes blacker. Above 700° C, the spectrum is almost featureless, and this indicates that all of the hydrogen has been driven off.

A major change in the f-QCC spectrum with increasing temperature is the large change in the $3.3-3.4 \mu m$ band ratio. At room temperature the $3.4 \mu m$ band is much stronger than the $3.3 \mu m$ band, and this indicates the dominance of saturated aliphatic C-H bonds. At 500° C the $3.3 \mu m$ band is stronger, and this indicates that the aromatic C-H bonds are dominant. Since the heating of f-QCC is done in vacuum, it is likely that the saturated C-H bonds that can be seen at $3.4 \mu m$ have been greatly reduced in the sample by loss of volatile components and by destruction of the saturated C-H bonds. The aromatic bonds that can be seen at $3.3 \mu m$ are not destroyed at 500° C and are present in the refractory component of f-QCC that remains in the heated sample.

Note that the QCC spectrum has peaks at 3.42 and 3.48 μ m and that it differs from the emission features seen in IRAS 21282+5050 and NGC 7027 at 3.46, 3.51, and 3.56 μ m (de Muizon *et al.* 1986; Nagata *et al.* 1988). We do not know whether QCC has any relationship to the emission features at 3.4–3.6 μ m, but it is possible that QCC is a member of a class of materials that gives rise to these emission features but QCC is not precisely the same material as that in the interstellar medium.

The heating of f-QCC is accompanied by a change in its appearance. Not only is the color changed, but inspection of f-QCC with an electron microscope shows a change in the appearance from a granulated filmy surface to spherical particles that are $0.1-10 \ \mu m$ in diameter. The temperature at which the change to spherical particles occurs is ~ 350° C. These effects—the change in the infrared spectrum, the shift of the peak absorbance of the 3.3 μm band, the change from a film to spherical particles—are referred to as the "thermal alteration" of f-QCC. The details of the thermal alteration of f-QCC will be discussed in a subsequent paper.

The most important result of this experiment is the shift of the peak absorbance of the 3.3 μ m feature from 3.289 to 3.295 μ m and the increased strength of the 3.3 μ m band compared to the 3.4 μ m band after heating to 500° C.

b) Comparison to PAH Spectra

The available spectra of PAHs in catalogs such as the Sadtler Handbook of Reference Spectra (Simons 1978) and the Aldrich Library of Infrared Spectra (Pouchert 1981) are inadequate for comparison to the astrophysical spectra, which have much higher spectral resolution. The high spectral resolution PAH spectra of Salisbury *et al.* (1988) are difficult to compare directly to the astrophysical spectra because the frequency scale is too compressed. Therefore the spectra of 20 PAHs were taken at the University of Electro-Communications with the same infrared spectrometer as used for obtaining the f-QCC spectra.

For the solid PAHs (powder or crystalline forms), the spectra were obtained by grinding a sample of PAH with KBr in a mortar and pressing the mixture into a pellet. This was then used to measure the infrared spectrum of the PAH. In the case of benzene, the spectrum was measured by grinding the 1990ApJ...353..543S

Number	Name	Formula	Number of Rings		PEAKS ^a (cm ⁻¹)	
1	Benzene	C ₆ H ₆	1	3036	3092	3072
2	Naphthalene	$\tilde{C_{10}H_8}$	2	3048	3063	3029
3	Anthracene	$C_{14}H_{10}$	3	3048	3310	3324
4	Phenanthrene	$C_{14}H_{10}$	3	3053	3045	3036
5	Naphthacene	$C_{18}H_{12}$	4	3044	3023	3058
6	Triphenylene	$C_{18}H_{12}$	4	3022	3058	3080
7	Pyrene	$C_{16}H_{10}^{12}$	4	3043	3029	2990
8	Benzanthracene	$C_{18}H_{12}$	4	3047	3030	3002
9	Chrysene	$C_{18}H_{12}$	4	3052	3020	3083
10	1,2,5,6-dibenzanthracene	$C_{2,2}H_{14}$	5	3033	3052	3071
11	Benzo[a]pyrene	$C_{20}H_{12}$	5	3033	3048	3074
12	Benzo[e]pyrene	$C_{20}H_{12}$	5	3046	3022	3077
13	1,2,3,4-dibenzanthrancene	$C_{22}H_{14}$	5	3053	3022	3076
14	Perylene	$C_{20}H_{12}$	5	3048		
15	Pentacene	$C_{22}H_{14}$	5	3041	3073	
16	3,4,8,9-dibenzopyrene	$C_{24}H_{14}$	6	3053	3028	
17	Benzoperylene	$C_{22}H_{12}$	6	3044	3012	
18	Coronene	$C_{24}H_{12}$	7	3018	3048	
19	Tetrabenzoperylene	$C_{34}H_{18}$	9	3042	3068	
20	Hexabenzocoronene	C_4 , H_{18}	13	3080	3070	3050

TABLE 1 ABSORBANCE PEAKS IN BENZENE AND PAHS

^a The three strongest absorbance peaks are listed in the order of strength.

KBr with liquid benzene, and a pellet was made as with the solid PAH. The spectrum was then measured quickly before the benzene could evaporate from the pellet. Effects of the KBr pellet on the infrared spectrum were investigated for benzene and found to be negligible (see below). The spectrum of gaseous benzene was also measured in a stainless steel gas cell with KRS-5 windows, to examine differences between the gas and liquid phase.

A summary of the PAH absorbance peaks we measured is given in Table 1 and plotted in Figure 2. Note that 15 out of 20 PAHs have a peak absorbance in the range 3040-3050 cm⁻¹ and that this is at a significantly higher frequency than the observed frequency of the interstellar emission feature (indicated by the dashed lines in Fig. 2). Among the 20 PAHs, only three, benzene, 1,2,5,6-dibenzanthracene, and benzo[a] pyrene, show a peak close to that of the interstellar emission. Also, the 3.3 μ m band of PAHs typically has a complex structure with multiple components (which probably arise from different fundamental vibrational modes), and it is broader than the interstellar emission band. Therefore a mixture of PAHs would produce a 3.3 μ m band that would be broader and more complex, and would probably peak at a higher frequency than the 3.29 μ m UIR emission feature.

A sample of such spectra (shown as the absorbance) as well as that of the thermally-altered f-QCC, is shown in Figure 3. The PAHs with absorbance maxima closest to that of the interstellar emission band at 3035 cm⁻¹ (3.295 μ m) are shown, as well as those of chrysene and coronene, which Allamandola, Tielens, and Barker (1985) and Léger and d'Hendecourt (1987) have used as representative interstellar PAHs. We also plot on a linear scale the emission spectrum of NGC 7027 that was previously obtained by Nagata et al. (1988). We chose a linear scale because the emission spectrum of NGC 7027 is likely to be optically thin and the absorbance is proportional to the absorption coefficient. Thus the spectrum of NGC 7027 and the absorbance spectrum should be directly comparable except for the uncertainty in the emission mechanism of the UIR emission features and particle size effects. Chrysene has a clear

peak at 3052 cm^{-1} , while coronene shows two peaks, at 3018and 3048 cm⁻¹; they are clearly different from the UIR 3.295 μ m (3035 cm⁻¹) emission feature. In summary, the thermally oxidized f-QCC shows the best resemblance to the UIR band.

The spectra of PAHs shown in Figure 3 were all taken in the solid phase and pressed into a KBr pellet. To understand the effect of the KBr pellet on the IR spectrum of PAHs, we measured another sample of benzene prepared in a different



FIG. 2.-The frequency of the peak absorbance of various samples of PAHs. Only the strongest peak is plotted. The numbers refer to the number of the PAH listed in Table 1. The dashed line indicates the frequency range within which the peak of the 3.29 μ m UIR emission feature is observed. Hexabenzocoronene (with 13 rings) has a peak at 3080 cm⁻¹ and is not shown.



FIG. 3.—The absorbance spectrum of oxidized f-QCC after heating to 500° C and PAHs compared to the emission spectrum of NGC 7027 obtained by Nagata *et al.* (1988). The Pf- δ line at 3.297 μ m has been removed from the spectrum of NGC 7027.

manner from that described previously. The sample was sandwiched between NaCl plates and measured. The resultant spectrum was completely identical to that in Figure 3. In addition, Léger, d'Hendecourt, and Défourneau (1989) show that wavelengths of the 3.3 μ m band of coronene measured in both the solid phase at 300 K and as isolated molecules in a solid neon matrix at 4.2 K differ by less than 1%. This is another indication that the matrix effects are small. The spectrum of f-QCC was taken with the sample deposited on a substrate; thus effects of the substrate should be negligible.

Differences between the gas phase and liquid phase spectra were also investigated for benzene and naphthalene. In benzene, the gas phase spectrum indicates fine structure due to rotational freedom and a slight shift (10 cm^{-1}) to higher frequencies in the peak absorption wavenumber. Also, multicomponents seen in the liquid phase absorption spectrum remains in the gas phase spectrum. A similar effect was observed for naphthalene. Thus it is likely that the disagreement between the PAH spectra and the 3.29 μ m interstellar feature pointed out above are true even for the gas phase PAH, and the gas phase spectrum cannot be expected to lead to a good match of the interstellar emission feature.

The spectra of PAHs so far presented are those for neutral and fully hydrogenated ones. In the interstellar medium, partially dehydrogenated and ionized species are expected and the excitation mechanism may be ultraviolet induced infrared fluorescence. The spectra of these species and under these conditions are not available at present and no detailed comparison between dehydrogenated/ionized PAHs and UIR emission features is possible. As far as the presently available data are concerned, thermally altered f-QCC gives the best fit to the 3.29 μ m feature.

III. DISCUSSION

a) What Is Thermally Altered Oxidized f-QCC?

The process for producing f-QCC is described by Sakata *et al.* (1987). It arises from thermally quenched C_2 and CH radicals from the same hydrocarbon plasma that are collected on the wall of the apparatus producing f-QCC. The production of the film occurs in a vacuum, and it is exposed to air to produce oxidized f-QCC. This oxidation process is essential to produce the 7.7 and 8.6 μ m bands in f-QCC. Since the f-QCC has many active sites, the oxidation proceeds rapidly.

To better understand the composition of f-QCC, the molecules and radicals from the hydrocarbon plasma were trapped on a cold finger at 20 K and were analyzed by mass spectroscopy. It was found to contain linear molecules (polyvnes), lowmolecular weight ring molecules (such as benzene and naphthalene), and their derivatives (Sakata 1980). In addition, more than 50% of the f-QCC is soluble in methanol. The dissolved material is low molecular weight material that is lost upon heating by evaporation. The trapped material is mainly molecular with a small amount of radicals. During heating from 20 K to room temperature, evaporation of material and further reactions such as polymerization caused by the radicals is expected. But the polymerization did not occur because the polymerization would have produced a dark residue, and this was not observed on the cold finger after heating. The molecules seen by the mass spectrometer are all low molecular weight rings, and this indicates that the basic unit in f-QCC is small rings (one and two rings).

The f-QCC was also mixed with deuterized chloroform $(CDCl_3)$, and proton NMR analysis of the soluble component indicates the presence of polyynes, ring molecules, olefinic molecules with double bonds, and methyl groups. The f-QCC is a condensate composed of these molecules and radicals. (Note: the CDCl₃ is a solvent which does not have hydrogen in it and is suitable for proton NMR.)

When f-QCC is heated, it is thermally altered. The heating causes (1) the loss of volatile low molecular weight components; (2) the destruction of some C-H bonds (dehydrogenation); and (3) the modification of C-C bonds (triple to double; double to single). The last two processes are a type of polymerization. The survival of the 3.3 and 11.4 μ m features after heating indicates the presence of the "solo" C-H struc-



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FIG. 4.-Examples of bridging between carbon rings.

ture in the thermally altered oxidized f-QCC in which carbon rings are connected through a bridge of carbon bonds (bridging) and these rings are randomly linked together in a three-dimensional structure. Examples of bridging are shown in Figure 4.

In the above figure, "rings" indicates one or more carbon rings, which are themselves linked to other groups. This "cross-linked" structure is different from the two-dimensional layered structure of graphite, and it is essential for producing the observed peak wavelength of 3.295 μ m in the thermally altered oxidized f-QCC.

b) Implications

The f-QCC forms naturally out of the hydrocarbon plasma, and its production is easily reproducible in the laboratory. There are three implications. First, the widespread distribution of the UIR emission features suggests to us that the method of producing the carbonaceous material should be relatively simple and should involve an abundant starting material. The process should also produce a very similar material throughout the Galaxy. The process producing f-QCC is consistent with this expectation. Although f-QCC is a "composite" with no well-defined chemical formula, it is a very specific material, since it is easily reproducible and matches the 3.29 μ m UIR emission feature in wavelength and FWHM. Second, the manner in which f-QCC is made indicates that the primary source of the carbonaceous material is likely to be the atmospheres of carbon-rich, mass-losing stars. This has been suggested as the source of the PAHs by Jura (1987), for example. The conditions of relatively high gas density, abundance of C₂ and CH, ~3000 K plasma temperature, and rapid cooling could be similar to that of an atmospheric parcel moving rapidly away from a star undergoing mass loss. Third, the nature of f-QCC is such that there are very few large PAHs in it; most of the aromatic molecules have one and at most two carbon rings. Therefore, if f-QCC is the carbonaceous material in space, then the formation of this material is less complex than that required for the formation of large PAHs with 40-60 carbon atoms. This fact helps to make it conceptually easier to see how the carbonaceous material can be so abundant and uniform in space.

Also, we note that the original motivation for producing QCC was to explain the 2175 Å ultraviolet feature, and it was concluded that QCC material (of the g-QCC type) can match the 2175 Å feature (Sakata et al. 1983). To our knowledge, the PAH hypothesis does not explain the ultraviolet extinction.

c) Comparison of the 3.29 Micron Band in f-QCC and PAHs

The comparison of the 3.29 μ m emission feature of PAHs and thermally altered oxidized f-QCC with that of the UIR emission features leads to the following conclusions:

First, the peak wavelength of the thermally altered oxidized f-QCC is precisely that of the 3.29 μ m UIR emission feature (3035 cm^{-1}) . This indicates the existence of a single type of C-H vibration mode in the thermally altered oxidized f-OCC. In contrast, most PAHs do not have a peak compatible with the observed UIR band, and in general the absorbance of large PAHs peaks at a shorter wavelength (higher frequency) than the 3.29 μ m UIR emission feature.

Second, the width of the thermally altered f-QCC is similar to that observed in NGC 7027, but it is much wider than that observed in HD 44179. A comparison is shown in Figure 5. The width of the 3.29 μ m absorption is a function of the degree of amorphousness (or disorder) and the spacing between the vibrating C-H units (or compactness). In principle, the width of the 3.29 μ m feature in f-QCC can be wider or narrower than what is shown in Figures 1 and 5, depending on these parameters of disorder and compactness. In contrast, the widths of the PAH 3.3 μ m absorbance, when the multiple components are taken into account, are much wider than the observed UIR band, and we do not see any possibility of obtaining narrower absorbance with PAHs. Thus, this is one of the major problems with the PAH hypothesis insofar as explaining the 3.29 μ m interstellar emission feature is concerned.

Third, the 3.29 μ m band of PAHs shows complex spectral structure, which results from the many vibrational modes in PAHs. Any PAH model for the UIR emission feature must address the problems of the generally poor frequency match, the narrowness, and the lack of structure of the UIR emission feature. For the above reasons, we think it is unlikely that the 3.295 μ m UIR emission feature can be assigned to a combination of isolated PAH molecules. The hypothesis that the 3.29 μ m UIR emission feature can result from a combination of large PAH molecules requires experimental verification. There is an additional problem of how to make the same specific mixture of PAH molecules under a wide range of astrophysical conditions. Of course, a similar problem exists for the f-QCC identification; however, we have shown that f-QCC can be made easily and reproducibly.



FIG. 5.-Comparison of the absorbance of thermally altered oxidized f-QCC to the emission spectrum of NGC 7027 and HD 44179. The Pf- δ line has been removed from the spectrum of NGC 7027, and a part of the spectrum of both NGC 7027 and HD 44179 near 3030 cm^{-1} has been removed because the strong telluric methane absorption prevents observations at this frequency.

IV. SUMMARY

New laboratory data on thermally altered f-QCC have been presented for direct comparison to astrophysical spectra of the 3.29 μ m UIR emission feature. This material, after heating to 500° C, has been altered by loss of volatiles, dehydrogenation, and polymerization. It has a "cross-linked" structure. In spite of its amorphous nature, thermally altered oxidized f-QCC can be made simply and reproducibly in the laboratory from a hydrocarbon plasma. The spectra of PAHs were also measured to make the same comparison. We find:

1. Thermally altered f-QCC has a single absorbance peak at 3.29 μ m that matches precisely the UIR emission feature.

2. The FWHM of the thermally altered f-QCC 3.29 μ m band is similar to that in NGC 7027, Orion, and IRAS 21282 + 5050.

3. Most PAHs do not have a peak absorbance that matches the UIR emission peak and the width is larger.

4. Most, if not all PAHs, show multiple absorption peaks

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near 3.29 μ m, in contrast to the single emission peak in the UIR emission feature.

For these reasons and from the comparison of spectral data (see Fig. 3) we conclude that thermally altered f-QCC provides a much better fit to the UIR 3.29 μ m emission feature.

In the comparison of oxidized f-QCC to the UIR emission features, we do not expect exact agreement on every detail of the UIR emission features, since it is impossible to duplicate the astrophysical conditions, time scales, and excitation mechanism in the laboratory. Based upon the precision of the wavelength match, the good fit to the width of the 3.29 μ m UIR emission feature, and the presence of features corresponding to the 3.4, 6.2, 7.7, 8.6, and 11.3 μ m features, we think that f-QCC is a good representative of the class of material giving rise to the UIR emission features.

We thank K. Sellgren for helpful comments. A. T. T. acknowledges the support of NASA contract NASW-3159.

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