The fullerene $C_{60}$ has four infrared-active vibrational transitions at 7.0, 8.5, 17.4, and 18.9 $\mu$m. We have previously observed emission features at 17.4 and 18.9 $\mu$m in the reflection nebula NGC 7023 and demonstrated spatial correlations suggestive of a common origin. We now confirm our earlier identification of these features with $C_{60}$ by detecting a third emission feature at 7.04 $\pm$ 0.05 $\mu$m in NGC 7023. We also report the detection of these three $C_{60}$ features in the reflection nebula NGC 2023. Our spectroscopic mapping of NGC 7023 shows that the 18.9 $\mu$m $C_{60}$ feature peaks on the central star and that the 16.4 $\mu$m emission feature due to polycyclic aromatic hydrocarbons peaks between the star and a nearby photodissociation front. The observed features in NGC 7023 are consistent with emission from UV-excited gas-phase $C_{60}$. We find that 0.1%–0.6% of interstellar carbon is in $C_{60}$; this abundance is consistent with those from previous upper limits and possible fullerene detections in the interstellar medium (ISM). This is the first firm detection of neutral $C_{60}$ in the ISM.

**Key words:** ISM: individual objects (NGC 7023, NGC 2023) – ISM: lines and bands – ISM: molecules – line: identification

## 1. INTRODUCTION

Fullerenes are cage-like molecules (spheroidal or ellipsoidal) of pure carbon such as $C_{60}$, $C_{70}$, $C_{76}$, and $C_{84}$, also known as buckminsterfullerene, is the most stable fullerene and can account for up to 50% of the mass of fullerenes generated in the laboratory (Kroto et al. 1985). Theorists have suggested that fullerenes might form around stars with carbon-rich atmospheres such as carbon stars, Wolf–Rayet (WC) stars, and carbon-rich, hydrogen-poor R Cr B stars (Kroto & Jura 1992; Goeres & Sedlmayr 1992; Cherchneff et al. 2000; Pascoli & Polleux 2000). Fullerenes may also form as part of the carbon-rich grain condensation process known to occur in the ejecta of Type II supernovae (Clayton et al. 2001). Hydrogenated amorphous carbon grains in the interstellar medium (ISM) may also decompose after interstellar shocks into polycyclic aromatic hydrocarbons (PAHs) and fullerenes (Scott et al. 1997); PAHs comprise 9%–18% of interstellar carbon (Joblin et al. 1992; Tielens 2008). Fullerenes might also form via cold interstellar gas-phase chemistry (Bettners & Herbst 1996, 1997).

Foing & Ehrenfreund (1994) propose that two diffuse interstellar bands at 958 and 963 nm are due to singly ionized $C_{60}$, or $C_{60}^+$, although the identification is debated (Maier 1994; Jenniskens et al. 1997). Misawa et al. (2009) attribute additional diffuse interstellar bands at 902, 921, and 926 nm to $C_{60}^-$. No fullerenes were found toward carbon-rich post-asymptotic giant branch stars (Somerville & Bellis 1989; Snow & Seab 1989), carbon stars (Clayton et al. 1995; Nuccitelli et al. 2005), or R CrB stars (Clayton et al. 1995; Lamb et al. 2001). Cami et al. (2010) have recently detected $C_{60}$ in the planetary nebula Tc 1 (IC 1266).

Observational evidence for neutral fullerenes in the ISM, however, has been elusive to date. No neutral fullerenes have yet been found in the diffuse ISM (Snow & Seab 1989; Herbig 2000), dense molecular clouds (Nuccitelli et al. 2005), or at the photodissociation front in the reflection nebula NGC 7023 (Moutou et al. 1999).

$C_{60}$ has four infrared-active vibrational transitions at 7.11, 8.55, 17.5, and 19.0 $\mu$m at 1200 K (gas phase; Frum et al. 1991), and at 6.98, 8.44, 17.3, and 18.9 $\mu$m at 2.4 K (para-H$_2$ matrix isolation; Sogoshi et al. 2000). On this basis, we tentatively identified the 17.4 and 18.9 $\mu$m ISM emission features in the reflection nebula NGC 7023 as due to $C_{60}$ (Werner et al. 2004b; Sellgren et al. 2007). We provide here additional evidence for the presence of $C_{60}$ in reflection nebulae. We report here the detection of the predicted $C_{60}$ feature at 7.04 $\pm$ 0.05 $\mu$m in NGC 7023. We also report the detection of $C_{60}$ features at 7.04, 17.4, and 18.9 $\mu$m in a second reflection nebula, NGC 2023. The $C_{60}$ 8.5 $\mu$m feature is too blended with strong 8.6 $\mu$m PAH emission to be detected in reflection nebulae. We have found in NGC 7023 (Sellgren et al. 2007) that the 16.4 $\mu$m emission feature, attributed to PAHs (Moutou et al. 2000), has a spatial distribution distinct from that of the 18.9 $\mu$m emission feature. We now compare the spatial distributions of the 16.4, 17.4, and 18.9 $\mu$m emission features in NGC 7023 and find additional support for the $C_{60}$ identification. We also discuss the excitation mechanism for the infrared emission of $C_{60}$.

## 2. OBSERVATIONS

We used the Spitzer Space Telescope (Werner et al. 2004a) with the Infrared Spectrograph (IRS; Houck et al. 2004) to obtain 5–38 $\mu$m spectra of NGC 2023 (PI: Sellgren, pid 40276, aorkeys = 23912704, 23911168) and NGC 7023 (PI: Sellgren, pid 40276, aorkeys = 23911424, 23911680). We obtained spectra with the short-wavelength low-resolution module SL (5–14 $\mu$m; $\lambda/\Delta\lambda = 60–120$), the long-wavelength low-resolution module LL (14–38 $\mu$m; $\lambda/\Delta\lambda = 60–120$), and the short-wavelength high-resolution module SH (9.5–19.5 $\mu$m;
Figure 1. *Spitzer*-IRS spectra (solid curves) of NGC 7023 (25" north, 4" east) and NGC 2023 (29" west, 8" south) with 5.2–10.0 μm and 10.0–130 μm resolution. (a) Short-wavelength low-resolution module (SL) and (b) short-wavelength high-resolution module (SH) spectra. The SL curves are plotted with solid lines and the SH curves with dashed lines. The blue curves show the observed spectra, the black curves show the fitted models, and the red curves show the residuals. The vertical lines at the edges of the plots indicate the wavelength range covered by each module.

Figure 2. *Spitzer*-IRS 5–9 μm spectrum of NGC 7023 (open squares), obtained with the short-wavelength low-resolution module (SL; 5.2–10.0 μm; λ/Δλ = 60–120) and the short-wavelength high-resolution module (SH; 10.0–19.5 μm; λ/Δλ = 600). We mark C₆₀ lines at 7.04, 8.5, 17.4, and 18.9 μm (vertical lines). The strong emission feature at 8.6 μm is due to PAHs. H₂ emission lines fall at 9.66, 12.7, and 17.0 μm.

λ/Δλ = 600). We chose nebular positions (29" west, 8" south of HD 37903 in NGC 2023; 25" east, 4" north of HD 200775 in NGC 7023) with a strong ratio of the 18.9 μm feature relative to the 16.4 μm PAH feature. We used matched aperture extraction in CUBISM (Smith et al. 2007b) to extract SL, LL, and SH spectra in regions of overlap between these spectral modules. The extraction aperture was 10′′ × 10′′ in NGC 2023 and 7′′ × 9′′ in NGC 7023.

We also retrieve from the *Spitzer* archive a spectral data cube for NGC 7023 with LL (PI: Joblin, pid 3512; arcsec = 0011057920). We use CUBISM to derive spectral images in the 16.4, 17.4, and 18.9 μm features and 0–0 S(1) H₂ for NGC 7023. For the spectrum of each spatial pixel, we define a local continuum surrounding an emission feature or line and subtract it before deriving the feature or line intensity.

We search for bad pixels and correct them with CUBISM before extracting final spectra. We subtract dedicated sky spectra for the 5–38 μm spectra of NGC 2023 and NGC 7023; no sky subtraction is done for the spectral mapping.

3. RESULTS

Figure 1 illustrates our SL and SH spectra in NGC 2023 and NGC 7023. The 17.4 and 18.9 μm emission features are prominent and coincident with C₆₀ wavelengths.

We show the 5–9 μm SL spectrum of NGC 7023 in Figure 2. We clearly detect an emission feature at 7.04 ± 0.05 μm. This feature is coincident, within the uncertainties, with the wavelength of the expected C₆₀ line. We highlight this emission feature by using PAHFIT (Smith et al. 2007a) to fit the 5–9 μm spectrum with a blend of PAH emission features in addition to the new emission feature at 7.04 μm. The full-width at half-maximum of the 7.04 μm C₆₀ feature is 0.096 ± 0.012 μm, significantly broader than our spectral resolution. We also detect the 7.04 μm C₆₀ feature in NGC 2023. We present the C₆₀ band intensities in Table 1.

In our previous long-slit spectroscopic investigation of NGC 7023 (Sellgren et al. 2007), we found that the 18.9 μm C₆₀ feature peaks closer to the central star than PAHs. We now illustrate this more clearly with the LL spectroscopic map extracted in NGC 7023 (Figure 3). The 18.9 μm emission is clearly centered on the star. By contrast, the 16.4 μm PAH emission peaks outside the region of maximum 18.9 μm emission, in a layer between the star and the molecular cloud. The photodissociation front at the UV-illuminated front surface of the molecular cloud is delineated by 0–0 S(1) H₂ emission at 17.0 μm.

Our previous observations (Sellgren et al. 2007) suggested that the 17.4 μm feature might be a blend of a PAH feature and an emission feature with the same spatial distribution as the 18.9 μm feature. We now confirm this is the case with IRS/LL spectroscopic imaging. We show an image of the 17.4 μm emission from NGC 7023 in Figure 4, overlaid with contours of 18.9 μm and 16.4 μm emission. The 17.4 μm emission clearly shows one peak on the central star, coincident with 18.9 μm C₆₀ emission, and a second peak cospatial with 16.4 μm PAH emission. Thus, there is an ISM component with emission features at 17.4 and 18.9 μm, which has a different spatial distribution than PAHs traced by the 16.4 μm feature.

Our imaging spectroscopy demonstrates the spatial separation between regions of peak PAH emission and peak C₆₀ emission.
HD 200775 (star). Each pixel is 5 μm. We adopt the absolute absorption cross-section at 0.09–0.30 μm in PAHs, we derive a percentage of interstellar carbon in C60, of 0.1%–0.6% from two infrared emission features. We do not include any potential contribution from C+ 60 (Yasumatsu et al. 1996; Yagi et al. 2009). The absorption cross-sections from Yasumatsu et al. (1996) and Yagi et al. (2009) differ by a factor of two, indicating the overall uncertainty in our abundance calculation. For PAHs, we adopt the absolute absorption cross-section at 0.09–0.30 μm from Li & Draine (2001). We integrated both of these absolute absorption cross-sections over a 17,000–22,000 K blackbody, as is appropriate for the central stars of NGC 7023 and NGC 2023. We find that C60 and PAHs have similar integrated UV absorption strengths per C atom.

We compare the sum of the intensities of the 7.04, 17.4, and 18.9 μm features, assumed to be due to C60, to the sum of all other infrared emission features at 5–38 μm, assumed to be due to PAHs. We analyze SL and LL spectra with PAHFIT (Smith et al. 2007a) to find that the ratio of C60 to PAH emission is 0.01–0.03 in our observed positions in NGC 7023 and NGC 2023. By adopting a percentage of interstellar carbon in PAHs of 9%–18%, we derive a percentage of interstellar carbon in C60, p(C60), of 0.1%–0.6% in regions of bright C60 emission.

Our p(C60) value is consistent with other estimates of p(C60) and of the percentage of carbon in C60, p(C60+). Foing & Ehrenfreund (1994) estimate p(C60+) = 0.3%–0.9% from two diffuse interstellar bands at 958 and 963 nm which they attribute to C60. Herbig (2000) uses these same two bands to estimate p(C60) = 0.1%–0.3% in diffuse clouds. Herbig (2000) places an upper limit of p(C60) < 0.0008% in diffuse clouds, showing that C60 is primarily ionized in diffuse clouds. Moutou et al. (1999) measure p(C60) < 0.3% and p(C60+) < 0.3% from the lack of emission features at 7.0–8.5 μm, at a position in NGC 7023 where the 18.9 μm C60 feature is weak. Nuccitelli et al. (2005) find p(C60) < 0.6% from their non-detection of the 8.5 μm feature in absorption toward R CrB (HR 5880) and three massive young stellar objects. Cami et al. (2010) derive p(C60) = 1.5% in planetary nebula Tc 1.

The relative intensities of the C60 bands provide information on the conditions in which the molecule emits. To probe these conditions, we use a Monte Carlo code, based on a micro-canonical formalism, developed to simulate the emission cascade of PAHs following the absorption of a UV photon (Joblin et al. 2002; Mulas et al. 2006). We calculate the evolution of the internal energy in the molecule and the number of photons emitted in each infrared band during the cooling cascade. We calculate the emission spectrum after C60 absorbs a 5, 10, or 15 eV photon, reaching temperatures of 800, 1200, or 1570 K at the beginning of the cooling cascade. We use the list of modes from Ménendez & Page (2000) and infrared intensities (at 0 K) calculated using density functional theory (Choi et al. 2000; Schettino et al. 2001). We compare the predicted line intensities with the observations in Table 1. We find that the ratio of the intensities of the 17.4 and 18.9 μm bands is not sensitive to the energy of the absorbed UV photon. The observed ratio varies but this is likely because the 17.4 μm band is blended with a PAH feature. The ratio of the intensities of the 7.04 and 18.9 μm bands, however, is very sensitive to the absorbed UV photon energy. Table 1 shows that the 7.04 μm intensity in NGC 7023 is consistent with the cooling cascade of C60 excited by UV photons with a mean energy of 10 eV. A lower photon energy is suggested for NGC 2023, perhaps because of its blister geometry with the star in front of a dense molecular cloud. If C60 in NGC 2023 is excited by lower energy photons, then this would also affect the C60 abundance derived there. More detailed modeling will be needed to clarify this.

Our interpretation of the NGC 7023 C60 emission differs from the very recent results of Cami et al. (2010) who concluded that

---

**Figure 3.** Three-color image of NGC 7023, in 18.9 μm C60 emission (red), 16.4 μm PAH emission (green), and 17.0 μm 0–0 S(1) H2 emission (blue). We constructed each image by analyzing Spitzer/IRS-LL long-slit spectra with CUBISM (Smith et al. 2007b). We illustrate where we measured the 5–38 μm emission. We mark the location of HD 200775 (star). Each pixel is 5′′ × 5′′. The 18.9 μm C60 feature peaks on the central star, while the 16.4 μm PAH emission is brightest between the 18.9 μm emission region and the photodissociation front traced by H2 emission.

**Figure 4.** Image of 17.4 μm emission in NGC 7023 (grayscale image), derived from Spitzer/IRS-LL long-slit spectra analyzed with CUBISM (Smith et al. 2007b). We overlay contours of 18.9 μm C60 emission (red) and 16.4 μm PAH emission (green). Each pixel is 5′′ × 5′′. One component of the 17.4 μm emission is co-spatial with 18.9 μm C60 emission and the other component of the 17.4 μm emission follows 16.4 μm PAH emission. This illustrates that the 17.4 μm emission feature is a blend of a PAH feature and 17.4 μm C60 emission.
C\textsubscript{60} in Tc 1 emits in solid phase at a temperature of 330 K. Their analysis, however, is based on an excitation diagram to derive a single emission temperature, which is not appropriate to describe the cooling cascade of UV-excited molecules.

5. CONCLUSIONS

We confirm our identification of the 17.4 and 18.9 $\mu$m emission features in NGC 7023 with C\textsubscript{60} (Werner et al. 2004b; Sellgren et al. 2007) by detecting a predicted emission feature at 7.04 $\pm$ 0.05 $\mu$m. We also detect 7.04, 17.4, and 18.9 $\mu$m emission features in NGC 2023. We demonstrate that the 17.4 $\mu$m emission feature in NGC 7023 is a blend of C\textsubscript{60} and PAH emission with Spitzer imaging spectroscopy. Our work (Werner et al. 2004b; Sellgren et al. 2007; this Letter) is the first firm detection of neutral C\textsubscript{60} in interstellar space.

We find that the infrared emission in NGC 7023 is consistent with the emission of gas-phase C\textsubscript{60} excited by UV photons. Further modeling is required to explain the observations of NGC 2023. The percentage of interstellar carbon in C\textsubscript{60} is 0.1%–0.6% in NGC 7023. This is consistent with previous estimates of, and limits on, the interstellar C\textsubscript{60} and C\textsuperscript{+}\textsubscript{60} abundances.

We thank Nick Abel, Lou Allamandola, Bruce Draine, Alain Léger, and Farid Salama for useful conversations, Dominique Toublanc for support with the Monte Carlo code, and Mike Jura for suggesting the C\textsubscript{60} identification. Our work (Werner et al. 2004b; Sellgren et al. 2007; this Letter) is the first firm detection of neutral C\textsubscript{60} in interstellar space.

Facilities: Spitzer (IRS)

REFERENCES

Bettens, R. P. A., & Herbst, E. 1996, ApJ, 468, 686
Bettens, R. P. A., & Herbst, E. 1997, ApJ, 478, 585
Boersma, C., Bauschlicher, C. W., Allamandola, L. J., Ricca, A., Peeters, E., & Tielens, A. G. G. M. 2010, A&A, 511, A32
Camí, J., Bernard-Salas, J., Peeters, E., & Malek, S. E. 2010, Science, 329, 1180
Cherchneff, I., Le Teuff, Y. H., Williams, P. M., & Tielens, A. G. G. M. 2000, A&A, 357, 572
Choi, C. H., Kertesz, M., & Mihaly, L. 2000, J. Phys. Chem. A, 104, 102
Clayton, D. D., Deneault, E. A.-N., & Meyer, B. S. 2001, ApJ, 562, 480
Clayton, G. C., Kelly, D. M., Lacy, J. H., Little-Marenin, I. R., Feldman, P. A., & Bernath, P. F. 1995, AJ, 109, 2096
Foing, B. H., & Ehrhardt, P. 1994, Nature, 369, 296
Frum, C. I., Engleman, R. J., Hedderich, H. G., Bernath, P. F., Lamb, L. D., & Huffman, D. R. 1991, Chem. Phys. Lett., 176, 504
Goeres, A., & Sedlmayr, E. 1992, A&A, 265, 216
Herbig, G. H. 2000, ApJ, 542, 334
Houck, J. R., et al. 2004, ApJS, 154, 18
Jenniskens, P., Mulass, G., Porceddu, L., & Benvenuti, P. 1997, A&A, 327, 337
Joblin, C., Léger, A., & Martin, P. 1992, ApJ, 393, L79
Joblin, C., Toublanc, D., Boissel, P., & Tielens, A. G. G. M. 2002, Mol. Phys., 100, 3595
Kroto, H. W., Heath, J. R., O’Brien, S. C., Curl, R. F., & Smalley, R. E. 1985, Nature, 318, 162
Kroto, H. W., & Jura, M. 1992, A&A, 263, 275
Lambert, D. L., Rao, N. K., Pandey, G., & Ivens, I. I. 2001, ApJ, 555, 925
Li, A., & Draine, B. T. 2001, ApJ, 554, 778
Maier, J. P. 1994, Nature, 370, 423
Ménendez, J., & Page, J. B. 2000, in Light Scattering in Solids VIII: Fullerene, Semiconductor Surfaces, Coherent Phonons, ed. M. Cardona & G. Güntherodt (Berlin: Springer), 27
Misawa, T., Gandhi, P., Hida, A., Tamagawa, T., & Yamaguchi, T. 2009, ApJ, 700, 1988
Moutou, C., Sellgren, K., Verstraete, L., & Léger, A. 1999, A&A, 347, 949
Moutou, C., Verstraete, L., Léger, A., Sellgren, K., & Schmidt, W. 2000, A&A, 354, L17
Mulass, G., Mallocc, G., Joblin, C., & Toublanc, D. 2006, A&A, 460, 93
Nuccitielli, D., Richter, M. J., & McCall, B. J. 2005, in IAU Symp. 235, Astrochemistry: Recent Successes and Current Challenges, Poster Book, ed. A. J. Markwick-Kemper (Cambridge: Cambridge Univ. Press), 236
Pascoli, G., & Polleux, A. 2000, A&A, 359, 799
Schettino, V., Pagliai, M., Ciabini, L., & Cardini, G. 2001, J. Phys. Chem. A, 105, 11192
Scott, A., Duley, W. W., & Pinho, G. P. 1997, ApJ, 489, L193
Sellgren, K., Uchida, K. I., & Werner, M. W. 2007, ApJ, 659, 1338
Smith, J. D. T., et al. 2007a, ApJ, 656, 770
Smith, J. D. T., et al. 2007b, PASP, 119, 1133
Snow, T. P., & Seab, C. G. 1989, A&A, 213, 291
Sogoshi, N., Kato, Y., Wakabayashi, T., Monose, T., Tam, S., DeRose, M. E., & Fajardo, M. E. 2000, J. Phys. Chem. A, 104, 3733
Somerville, W. B., & Bellis, J. G. 1989, MNRS, 240, 41P
Tielens, A. G. G. M. 2008, ARA&A, 46, 289
Velusamy, T., & Langer, W. D. 2008, ApJ, 136, 602
Werner, M. W., Uchida, K. I., Sellgren, K., Marengo, M., Gordon, K. D., Morris, P. W., Houck, J. R., & Stansberry, J. A. 2004b, ApJS, 154, 209
Werner, M. W., et al. 2004a, ApJS, 154, 1
Yagi, H., et al. 2009, Carbon, 47, 1152
Yasumatsu, H., Kondow, T., Kitagawa, H., Tabayashi, K., & Shobatake, K. 1996, J. Chem. Phys., 104, 899