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On buckyonions as an interstellar grain component

Aigen Li,[1,2]★ J. H. Chen,[1,2]★ M. P. Li,[2]★ Q. J. Shi[1,3]★ and Y. J. Wang[1]★

[1]*Department of Physics, Hunan Normal University, Changsha, Hunan 410081, China*
[2]*Department of Physics and Astronomy, University of Missouri, Columbia MO 65211, USA*
[3]*Department of Science and Engineering of Shuda College, Hunan Normal University, Changsha, Hunan 410081, China*

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ABSTRACT

The carrier of the 2175 Å interstellar extinction feature remains unidentified since its first detection over 40 yr ago. In recent years, carbon buckyonions have been proposed as a carrier of this feature, based on the close similarity between the electronic transition spectra of buckyonions and the 2175 Å interstellar feature. We examine this hypothesis by modelling the interstellar extinction with buckyonions as a dust component. It is found that dust models containing buckyonions (in addition to amorphous silicates, polycyclic aromatic hydrocarbon molecules, graphite) can closely reproduce the observed interstellar extinction curve. To further test this hypothesis, we call for experimental measurements and/or theoretical calculations of the infrared vibrational spectra of hydrogenated buckyonions. By comparing the infrared emission spectra predicted for buckyonions vibrationally excited by the interstellar radiation with the observed emission spectra of the diffuse interstellar medium, we will be able to derive (or place an upper limit on) the abundance of interstellar buckyonions.

Key words: dust extinction – ISM: lines and bands – ISM: molecules.

1 INTRODUCTION

In the interstellar medium (ISM), the strongest spectroscopic extinction feature is the 2175 Å bump which is characterized with a stable peak wavelength and a width variable from one sightline to another (Fitzpatrick & Massa 2007). Since Stecher & Donn (1965) first detected this ultraviolet (UV) extinction feature through rocket observations, the origin of this feature and the nature of its carrier(s) are still an enigma. Many candidate materials, including graphite, amorphous carbon, graphitized (dehydrogenated) hydrogenated amorphous carbon, nano-sized hydrogenated amorphous carbon, quenched carbonaceous composite, coals, polycyclic aromatic hydrocarbon (PAH), and OH^- ion in low-coordination sites on or within silicate grains have been proposed, while no single one is generally accepted (see Li & Greenberg 2003 for a review).

Recently, Chhowalla et al. (2003) measured the UV-visible photoabsorption spectra of carbon buckyonions (BOs)[1] composed of spherical concentric fullerene shells (so far the largest BOs produced in laboratory have $N \sim 100$ shells; Iglesias-Groth et al. 2003). They found that the plasmon-like feature of BOs due to a collective excitation of the π electrons closely fits the 2175 Å interstellar extinction feature. More recently, Ruiz, Bretón & Gomez Llorente (2005) theoretically simulated the photoabsorption spectra of BOs.[2] They found that the calculated absorption spectra of BOs are in close agreement with the experimental data of Chhowalla et al. (2003) and the observed 2175 Å interstellar feature. That the π-plasmon absorption band of BOs exhibits a stable peak position at ∼5.70 eV ($\lambda \approx 2175$ Å, $\lambda^{-1} \approx 4.6\,\mu\text{m}^{-1}$) and a variable bandwidth (Chhowalla et al. 2003; Ruiz et al. 2005) suggests that BOs may be a promising candidate material for the 2175 Å interstellar extinction feature.

Indeed, it is shown by Chhowalla et al. (2003) and Ruiz et al. (2005) that the photoabsorption spectra of BOs *alone* very well reproduce the entire interstellar extinction curve at $\lambda^{-1} \sim 3.2$–$7.3\,\mu\text{m}^{-1}$. This requires ∼190 ppm (parts per million) C/H to be locked up in BOs (Ruiz et al. 2005). However, it is well recognized that in the interstellar medium (ISM), in addition to the 2175 Å extinction carrier, there must exist other dust components as well – there must be a population of amorphous silicate dust, as indicated by the strong, ubiquitous 9.7 and 18 μm interstellar absorption features; there must be a population of aromatic hydrocarbon dust (presumably PAH molecules), as indicated by the distinctive set of 'unidentified' infrared (UIR) emission bands at 3.3, 6.2, 7.7,

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★E-mail: lia@missouri.edu (AL); limo@missouri.edu (MPL); jhchen@hunnu.edu.cn (JHC); qjshi@hunnu.edu.cn (QJS); wyj@hunnu.edu.cn (YJW)

[1] Early pioneering experimental studies of the UV absorption properties of BOs and their association with the 2175 Å interstellar extinction feature include those of de Heer & Ugarte (1993), Ugarte (1995), and Wada et al. (1999).

[2] Early pioneering theoretical studies of the UV absorption properties of BOs and their association with the 2175 Å interstellar extinction feature include those of Wright (1988), Henrard, Lucas & Lambin (1993), Henrard et al. (1997) and Lucas, Henrard & Lambin (1994).

8.6 and 11.3 μm ubiquitously seen in the ISM; there must also exist a population of aliphatic hydrocarbon dust, as indicated by the 3.4 μm C–H absorption feature which is also ubiquitously seen in the diffuse ISM of the Milky Way and external galaxies (see Li 2004).

Although BOs are able to closely reproduce the 2175 Å extinction feature, it is not clear if dust models with amorphous silicates, PAHs and other carbon dust species (e.g. amorphous carbon, hydrogenated amorphous carbon, organic refractory and graphite) incorporated (in addition to BOs) are still capable of fitting the 2175 Å extinction feature. One may intuitively expect that the almost perfect fit to the $\lambda^{-1} \sim 3.2$–$7.3\ \mu\mathrm{m}^{-1}$ interstellar extinction would easily be distorted by the addition of other dust components. It is the purpose of this *Letter* to examine this issue. To this end, we consider the extinction of dust models consisting of multimodal grain populations with BOs as an interstellar grain component.

2 MODELS

We consider dust models consisting of amorphous silicate, graphite, PAHs and BOs. We take the size distributions of silicate and graphite dust to be either that of Mathis, Rumpl & Nordsieck (1977, hereafter MRN) or Weingartner & Draine (2001, hereafter WD). The MRN size distribution is a simple power law $\mathrm{d}n/\mathrm{d}a \sim a^{-3.5}$ with a lower cut-off at $a_{\mathrm{min}} = 50$ Å and an upper cut-off at $a_{\mathrm{max}} = 0.25\ \mu$m for both silicate and graphite. The WD size distribution is more extended. Unlike the MRN distribution, it smoothly extends from $a_{\mathrm{min}} = 3.5$ Å up to ∼1 μm for both silicate and graphite.[3] We also include a PAH component with a lognormal size distribution $\mathrm{d}n/\mathrm{d}\ln a \propto \exp\{-[\ln(a/a_{\mathrm{o}})]^2/(2\sigma^2)\}$ for $a \geq 3.5$ Å, where $a_{\mathrm{o}} = 3.5$ Å and $\sigma = 0.4$ determine the peak location and width of the distribution (Li & Draine 2001; WD). For BOs, we do not need to consider size distributions since the UV-visible photoabsorption spectra are practically independent of size for BOs with the number of nested shells $N > 3$ (Chhowalla et al. 2003; Ruiz et al. 2005).

The quantity of dust is taken to be consistent with the interstellar abundance constraints: the dust-forming elements (C, O, N, Si, Mg, Fe), if not seen in the gas phase, must have been depleted into dust. This requires an accurate knowledge of the interstellar reference abundance (i.e. the total abundance of an element both in gas and dust).

Let $[\mathrm{X/H}]_{\mathrm{ism}}$ be the interstellar reference abundance of element X relative to H, $[\mathrm{X/H}]_{\mathrm{gas}}$ be the gas-phase abundance of X (relative to H). The abundance of X (relative to H) in dust is $[\mathrm{X/H}]_{\mathrm{dust}} = [\mathrm{X/H}]_{\mathrm{ism}} - [\mathrm{X/H}]_{\mathrm{gas}}$. We assume that all cosmically available Si, Mg and Fe are locked up in amorphous silicate dust, i.e. $[\mathrm{Si/H}]_{\mathrm{dust}} = [\mathrm{Si/H}]_{\mathrm{ism}}$, $[\mathrm{Mg/H}]_{\mathrm{dust}} = [\mathrm{Mg/H}]_{\mathrm{ism}}$ and $[\mathrm{Fe/H}]_{\mathrm{dust}} = [\mathrm{Fe/H}]_{\mathrm{ism}}$ (see Savage & Sembach 1996). We take $[\mathrm{C/H}]_{\mathrm{gas}} = 140$ ppm (Cardelli et al. 1996).

We assume the interstellar reference abundance to be solar, i.e. $[\mathrm{X/H}]_{\mathrm{ism}} = [\mathrm{X/H}]_{\odot}$ ($[\mathrm{C/H}]_{\mathrm{ism}} = [\mathrm{C/H}]_{\odot} \approx 361$ ppm, $[\mathrm{Si/H}]_{\mathrm{ism}} = [\mathrm{Si/H}]_{\odot} \approx 35$ ppm, $[\mathrm{Mg/H}]_{\mathrm{ism}} = [\mathrm{Mg/H}]_{\odot} \approx 36$ ppm, $[\mathrm{Fe/H}]_{\mathrm{ism}} = [\mathrm{Fe/H}]_{\odot} \approx 30$ ppm).[4]

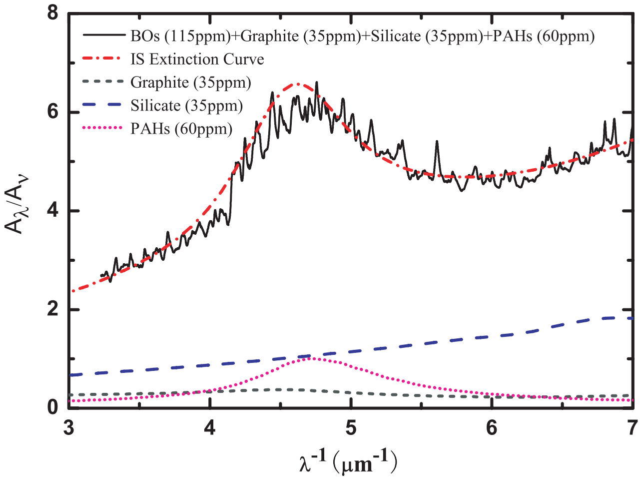

Figure 1. Comparison of the interstellar extinction (dot–dashed line) with the model extinction (solid line with oscillatory features) obtained by summing up the contributions of BOs (C/H = 115 ppm), PAHs (C/H = 60 ppm), graphite (C/H = 35 ppm) and amorphous silicate (Si/H = 35 ppm). We assume a MRN-type size distribution for the silicate and graphite grains.

With $[\mathrm{C/H}]_{\mathrm{gas}} = 140$ ppm in the gas phase, we have $[\mathrm{C/H}]_{\mathrm{dust}} \approx 221$ ppm left for carbon dust: BOs, PAHs and graphite. The 'UIR' bands require C/H ≈ 60 ppm to be in PAHs (Li & Draine 2001). So, we have C/H = 161 ppm for BOs and graphite. For silicate dust, we assume a stoichiometric composition of $\mathrm{MgFeSiO_4}$ and Si/H = Mg/H = Fe/H = 35 ppm.

With these parameters specified, we just vary the amounts of C (relative to H) depleted in BOs and graphite to fit the interstellar extinction curve. As shown in Fig. 1, with C/H = 35 ppm in graphite and C/H = 115 ppm in BOs, the dust model consisting of multimodal grain populations and with a MRN-type size distribution closely fits the interstellar extinction curve at $\lambda^{-1} \sim 3.2$–$7\ \mu\mathrm{m}^{-1}$ (at present the photoabsorption spectra of BOs are available only in this wavelength range). This model requires C/H = 210 ppm smaller than the total available $[\mathrm{C/H}]_{\mathrm{dust}} = 221$ ppm. The extinction cross-sections of amorphous silicate and graphite are calculated from Mie theory [using the dielectric functions of Draine & Lee (1984) and assuming a spherical shape for these grains]. The absorption spectra of PAHs and BOs are taken, respectively, from Draine & Li (2007) and Ruiz et al. (2005). The latter was an average of eight BOs for which the number of nested shells $N = 3, 4, \ldots, 10$ (see Ruiz et al. 2005). Note that the UV-visible absorption spectra of BOs are essentially independent of N (Ruiz et al. 2005).

Similarly, we have considered a WD-type size distribution for the silicate and graphite dust components. As shown in Fig. 2, with C/H = 20 ppm in graphite and C/H = 130 ppm in BOs, the multicomponent grain model provides a reasonably close match to the interstellar extinction curve. The total required carbon abundance is also C/H = 210 ppm, smaller than the total available $[\mathrm{C/H}]_{\mathrm{dust}} = 221$ ppm.

The oscillatory features in the BO model spectra arise from the electronic transitions of BOs. As stated in Ruiz et al. (2005), not all the oscillations in the photoabsorption spectra of BOs are reliable, but the general features such as the average width and peak position are quite robust and reliable. If BOs are indeed present in the ISM, we would expect a mixture of BOs of various sizes and various degrees of hydrogenation, ions and radicals of which the oscillatory features occur at different wavelengths; therefore, as a

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[3] The optical properties of graphite are taken to be that of PAHs at very small sizes ($a \leq 25$ Å) and that of graphite at radii $a \geq 100$ Å (see Li & Draine 2001).

[4] The published solar abundances have undergone major changes over the years and are still subject to major systematic uncertainties, as demonstrated in table 1 of Li (2005). We take the average values of the widely used solar abundances compiled by Grevesse & Sauval (1998) and Holweger (2001).

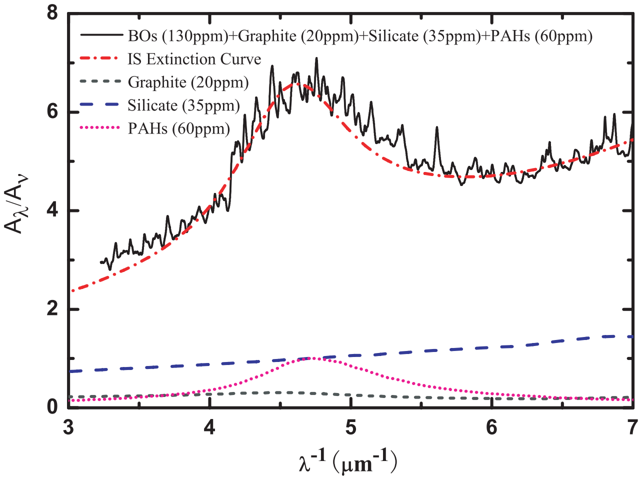

Figure 2. Same as Fig. 1 but with a WD-type size distribution assumed for the silicate and graphite grains.

collective effect, the strong indentations seen both experimentally and theoretically in the photoabsorption spectra of individual BOs will likely be smoothed out and will not show up in astronomical spectra.

3 DISCUSSION

It is encouraging that the dust models consisting of multimodal grain populations (including BOs) fit the interstellar extinction curve (including the 2175 Å extinction feature) very well while satisfying the interstellar abundance constraints. The fit to the 2175 Å feature is mainly affected by the silicate dust component. The smaller amount of silicate dust is included in the model, the better is the fit. We would achieve a closer match to the 2175 Å extinction if we take the interstellar Si abundance to be subsolar (Snow & Witt 1996) like that of B stars $[\mathrm{Si/H}]_{\mathrm{dust}} = 18$ ppm (Sofia & Meyer 2001) while having $\mathrm{C/H} = 140$ ppm in BOs and $\mathrm{C/H} = 10$ ppm in graphite.[5]

BOs are a promising candidate material for the 2175 Å interstellar extinction feature: (i) their π-plasmon absorption profile closely resembles the 2175 Å extinction feature and is stable in peak wavelength position while varies in width;[6] (ii) the almost perfect fit to the interstellar feature is not significantly distorted by the inclusion of other dust components (e.g. silicate, PAHs, graphite) which are required to account for other interstellar phenomena (e.g. the 9.7, 18 μm absorption features, the 'UIR' bands) and (iii) BOs are highly stable molecules that they can survive under intense UV radiation and are highly resistant to destruction by collisions.

BOs can be formed by electron beam irradiation of carbon soot (Ugarte 1992) and by heat treatment (annealing at temperatures $T \geq 700^\circ$C) and electron beam irradiation of nanodiamond (Kuznetsov et al. 1994; Tomita, Fujii & Hayashi 2002). The generation of BOs by annealing nanodiamonds is astrophysically relevant. Presolar nanodiamonds are identified in primitive carbonaceous meteorites based on their isotopic anomalies (Lewis et al. 1987). The possible existence of nanodiamonds in dense clouds and circumstellar dust discs or envelopes have also been suggested (Allamandola et al. 1992; Guillois, Ledoux & Reynaud 1999; van Kerckhoven, Tielens & Waelkens 2002). The generation of BOs through annealing nanodiamonds can occur in the ISM where nanodiamonds are stochastically heated to temperatures as high as ∼1000 K by the interstellar radiation field (Jones & d'Hendecourt 2000). Indeed, BOs have been found in meteorites with anomalous isotopic compositions (Smith & Buseck 1981; Bernatowicz et al. 1996; Harris, Vis & Heymann 2000).

Recently, BOs have also been invoked to explain other interstellar phenomena: the mysterious diffuse interstellar bands (DIBs) resulting from the electronic transitions of BOs (Iglesias-Groth & Bretón 2000; Iglesias-Groth 2004),[7] the 10–100 GHz Galactic anomalous microwave emission resulting from the electric dipole emission of rotationally excited BOs (Iglesias-Groth & Bretón 2000; Iglesias-Groth 2005), and the broad feature around 100 μm of the diffuse emission from two active star-forming regions (the Carina Nebula and Sharpless 171) resulting from the small particle surface resonance (Onaka & Okada 2003).

Experimentally, all BOs are found to belong to the C_{60N^2} icosahedral family with $N = 1, 2, \ldots$ (i.e. their shells are consecutive elements of the icosahedral C_{60N^2} fullerene family, with C_{60} being the smallest shell[8] and the intershell distance being very close to the interplanar separation in graphite, often taken to be 3.55 Å, the C_{60} radius; Yoshida & Osawa 1993; Ruiz, Bretón & Gomez Llorente 2004). So far, all the experimentally generated and analysed BOs are in the nanometer size range (∼3–15 nm in diameter; see de Heer & Ugarte 1993; Cabioc'h et al. 1997; Chhowalla et al. 2003).

The fullerenes produced in laboratory are usually accompanied by tubular and other non-spherical graphite-like structures of which the theoretical UV spectra exhibit some similarity to that of BOs. In the ISM, tubular graphite and PAHs can be catalytically formed at $T \geq 1000$ K on Fe nanoparticles and are probably the carrier of

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[5] If both C and Si are subsolar (say, two-third of that of solar, see Snow & Witt 1996), we will not be able to fit the interstellar extinction (also see Li 2005).

[6] Ruiz et al. (2005)'s theoretically simulated photoabsorption spectra of BOs showed that the width of the 2175 Å feature γ_{bump} varies with the size for small BOs, however, the width becomes independent of size when the number of shells $N \geq 6$ ($\gamma_{\mathrm{bump}} \approx 1.03\ \mu\mathrm{m}^{-1}$; see their fig. 4). For BOs to explain the observed 2175 Å interstellar features broader than $\gamma_{\mathrm{bump}} = 1.03\ \mu\mathrm{m}^{-1}$ (some sightlines have γ_{bump} up to $\sim 1.2\ \mu\mathrm{m}^{-1}$), clustering of individual BOs (de Heer & Ugarte 1993; Rouleau, Henning & Stognienko 1997), imperfect growth of BOs (e.g. mixing with amorphous carbon impurities; see Chhowalla et al. 2003) and coating BOs with a layer of PAHs (Mathis 1994) may play an important role.

[7] To date, >300 DIBs have been detected in the Galactic and extragalactic ISM. These mysterious optical to near-IR absorption lines (broader than the narrow lines from gas atoms, ions and small molecules) still remain unidentified. Fullerenes are known to have strong structural resonances at this wavelength range (Dresselhaus, Dresselhaus & Eklund 1996; Iglesias-Groth 2004). Iglesias-Groth (2007) argued that BOs may be responsible for the strongest optical DIB at 4430 Å and possibly also the 6177 and 6284 Å DIBs.

[8] Kroto et al. (1985) first proposed that C_{60} could be present in the ISM with a considerable quantity. This molecule and its related species were later proposed as the carriers of the 2175 Å extinction hump, the DIBs, the 'UIR' bands and the extended red emission (see Webster 1991, 1992, 1993a,b). C_{60} and C_{70} are unlikely a major contributor to the 2175 Å extinction feature since they have a characteristic doublet absorption in this wavelength region. Foing & Ehrenfreund (1994) attributed the two DIBs at 9577 and 9632 Å to C_{60}^{+}. However, attempts to search for these molecules in the UV and IR were unsuccessful (Snow & Seab 1989; Somerville & Bellis 1989; Moutou et al. 1999; Herbig 2000). These molecules are now estimated to consume at most <0.7 ppm carbon (Moutou et al. 1999). Therefore, C_{60} is at most a minor component of the interstellar dust family.

some of the DIBs (Zhou et al. 2006). They may also contribute to the 2175 Å interstellar extinction feature.

Finally, we should note that a powerful test of the BOs hypothesis would be in the IR. Because of their small sizes (and therefore small heat capacities), in the ISM, BOs will be stochastically heated by single UV photons (Draine & Li 2001). With their surface shell hydrogenated, they will emit in the near- to mid-IR through their characteristic C–H stretching and bending bands, and C–C stretching bands. The detection (or non-detection) of these bands will allow us to derive (or place an upper limit on) the abundance of BOs. Note that the BO models require an appreciable amount of C/H to be in BOs (>110 ppm), nearly twice as much as PAHs. Unfortunately, little is known about the positions and strengths of these vibrational bands. We call for urgent experimental measurements and/or theoretical calculations of the IR vibrational spectra of hydrogenated BOs. Future far-IR/submm space missions (e.g. Herschel) will also be useful for studying their far-IR vibrational spectra (see Iglesias-Groth & Bretón 2000). Moreover, it would be interesting to see if the silicate–graphite–PAH–BO multicomponent dust model is able to reproduce the $\sim$2–3000 μm overall IR emission of the Galactic ISM.

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