

# Interstellar diamonds in the Murchison meteorite: Isotopic characterization

S. V. S. Murty, G. Srinivasan and J. N. Goswami

Physical Research Laboratory, Navrangpura, Ahmedabad 380 009, India

Interstellar microdiamonds, typically 15–30 Å in size, are present in primitive meteorites. The extra-solar origin of these diamonds is confirmed by the presence of isotopic anomalies in nitrogen and xenon. We have successfully isolated microdiamonds from the Murchison meteorite and confirmed their interstellar origin based on their isotopic composition. Observation of neon isotopic anomaly characterized by an almost pure  $^{22}\text{Ne}$  component suggests the presence of a small amount of fine-grained interstellar silicon carbide in the analysed sample. Plausible scenarios for the origin of interstellar microdiamond that are consistent with the isotopic data are presented. The presence of interstellar microdiamonds in meteorites poses a challenge to the astronomers as direct observational evidence for the presence of diamond in interstellar space is yet to be found.

THREE distinct types of carbon-bearing phases of interstellar origin (diamond, graphite and silicon carbide) have been discovered in several primitive meteorites in recent years<sup>1–3</sup>. The extra-solar origin of these phases has been proven by the presence of isotopic anomalies (departure from normal solar system isotopic abundances) in carbon, nitrogen, silicon, noble gases and several other elements. The abundance of these three phases varies from one meteorite to another. Diamond is the most abundant phase, amounting up to a few hundred ppm (parts per million) of the whole meteorite. Silicon carbide and graphite are present only at ppm level. A brief summary of the salient features of the interstellar phases found in primitive meteorites is given in Table 1. The most widely studied interstellar phases in meteorites are microdiamonds and silicon carbide (SiC). The average size of 15–30 Å for diamond precludes its analysis individually, while SiC grains with maximum sizes of up to several microns can be analysed individually by ion microprobe, laser-probe mass spectrometer and transmission electron microscope<sup>4–6</sup>. The isotopic anomalies in C, Si, N and noble gases in SiC (e.g. overabundance of slow neutron capture (s-process) nucleosynthetic product ( $^{128}\text{Xe}$ ), enriched abundances of  $^{13}\text{C}$  (relative to  $^{12}\text{C}$ ) and of  $^{29}\text{Si}$  and  $^{30}\text{Si}$  (relative to  $^{28}\text{Si}$ )), have established that most of the SiC grains were formed in the envelopes of carbon

Table 1. Some characteristics of interstellar grains identified in primitive meteorites\*

	Diamond	Silicon carbide	Graphite
Abundance (ppm)**	~ 400	~ 5	~ 1–5
Size (µm)	≤ 0.003	< 0.1–10	< 0.1–6
Anomalous isotopic component <sup>†</sup>	Xe-HL	Xe-S, Ne-E	Ne-E
Associated isotopic component <sup>†</sup>	Ne-A2, Xe-P	Kr-S	—
$\delta^{13}\text{C}$ (per mil)	-38	600–6000	~ 350
$\delta^{15}\text{N}$ (per mil)	-330	< -400	> 250

\*Source of Data: Anders<sup>29</sup>.

\*\*The abundance varies from meteorite to meteorite. The quoted values are for the Murchison meteorite.

<sup>†</sup> See the Results section for the definitions of noble gas components.

stars during their asymptotic giant branch (AGB) phase of evolution<sup>7–9</sup>. The most characteristic signatures of the interstellar microdiamonds isolated from meteorites are the presence of a distinct isotopic anomaly in nitrogen with a depletion of  $^{15}\text{N}$  (the nitrogen isotopic ratio,  $^{15}\text{N}/^{14}\text{N}$ , is ~ 30% below the atmospheric value)<sup>10</sup> and a xenon component that shows clear enrichment in both the light ( $^{124}, ^{126}\text{Xe}$ ) and heavy ( $^{134}, ^{136}\text{Xe}$ ) isotopes compared to solar abundances<sup>11</sup>. The relative abundances of nitrogen and xenon in interstellar diamonds are however very different. The nitrogen abundance is quite high, ~ 2000–10,000 ppm, resulting from the fact that nitrogen can substitute carbon in the tetrahedral lattice of diamond, whereas the xenon abundance is extremely low, a few parts per billion (ppb). One can therefore consider nitrogen in diamond to be cogenetic and use it as a tracer for obtaining information on the source and origin of interstellar diamond. On the other hand, the anomalous xenon isotopic signal could be imprinted in diamonds either at their source region or during their interstellar history. Even though several research groups have carried out studies of nitrogen and noble gases in interstellar diamonds in recent years<sup>10–13</sup>, these were not analysed simultaneously because of instrumental limitations. Thus, a possible association between xenon and nitrogen isotopic systematics in interstellar microdiamonds has been inferred from data obtained in separate experiments. We have recently upgraded a commercial all-metal noble gas

mass spectrometer (VG 1200) through indigenous effort for simultaneous analysis of nitrogen and noble gases<sup>14, 15</sup>. This allowed us to perform the first simultaneous analysis of isotopic compositions of these elements in microdiamonds isolated from the carbonaceous chondrite Murchison. Here we present the results obtained in this study, which indicate a clear association between nitrogen and xenon isotopic anomalies in interstellar microdiamonds. We also consider the implications of the results for the source and origin of interstellar microdiamonds and their subsequent interstellar history before they became a part of the nascent solar nebula ~ 4.6 billion years ago.

### Experimental approach

The discovery of interstellar grains in meteorites resulted from more than a decade of sustained effort to isolate the carrier phase(s) of noble gas isotopic anomalies found in several primitive meteorites. The basic procedure adopted was to destroy the more common silicate, metal, sulphide and organic phases in meteorites by a suitable chemical treatment, and analyse individual components of the residue, comprising primarily refractory oxides and carbon-bearing phases, to pinpoint the plausible carrier phase(s) of isotopic anomalies. The carbon-bearing phases were of particular interest because graphite and silicon carbide are well-known components of interstellar dust. Furthermore, thermodynamic considerations suggest that these phases can be produced only in carbon-rich environment ( $C/O \geq 1$ )<sup>16</sup> and as such they could not have formed in the early solar system environment with  $C/O \sim 0.5$ . Earlier studies<sup>1-3, 10, 11</sup> have shown that three different carbon-bearing phases – microdiamond, SiC and graphite – can be recovered from chemically resistant residues of primitive meteorites; all these phases have isotopic anomalies that confirm their extra-solar (interstellar) origin. In the present experiment we have followed a particular approach<sup>17</sup> to isolate microdiamonds from the carbonaceous chondrite Murchison. The choice of the meteorite was based on the fact that carbonaceous chondrites are some of the most primitive meteorites and earlier studies have revealed the presence of interstellar microdiamonds in them at measurable levels (see Table 1). A brief description of the experimental procedure is given below.

About 6 g of the Murchison meteorite was crushed to ~ 100  $\mu\text{m}$  size in an agate mortar. It was treated with a mixture of HF + HCl and 6 N HCl alternatively in a teflon beaker. Five such cycles, each lasting nearly 24 h, were carried out until no change in the residual mass was noticed. This operation was to destroy the silicate minerals and to liberate any refractory and

acid-resistant grains trapped inside them. Sulphides that could be present were destroyed by treating with  $\text{CS}_2$ . The residue at this stage consisted mainly of carbonaceous matter (including organic matter) and refractory oxide minerals (e.g. spinel). Treatment with concentrated  $\text{HNO}_3$  at 160°C and then with  $\text{HClO}_4$  at boiling point was carried out to oxidize completely the organic matter, leaving behind a residue that consisted primarily of refractory carbonaceous matter. A major fraction of this residue is expected to be diamond with a small admixture of graphite and SiC. Diamond was separated from this mixture in the form of a colloidal suspension by treating with  $\text{NH}_3$  solution. Microdiamonds were recovered by acidifying the colloid. The identification and extra-solar origin of the separated microdiamonds are based primarily on the results of isotopic investigations, discussed in the next section. The abundance of diamond in the sample was a few hundred ppm.

An aliquot of the separated diamond sample, weighing about 1.5 mg, was analysed in the upgraded VG 1200 all-metal mass spectrometer to determine nitrogen and noble gas isotopic compositions. The upgradation of the instrument involved mainly the designing of a special extraction system that is capable of processing nitrogen and noble gases simultaneously<sup>15</sup>. Using this system, one can extract gases by either combustion or pyrolysis, and in both procedures the nitrogen blanks are very low (a few nanograms) and allow high-precision measurements of isotopic compositions of nitrogen and noble gases in small samples ( $\leq \text{mg}$ ). The microdiamond sample from Murchison was wrapped in a gold foil and loaded into the extraction system of the mass spectrometer. The sample was heated up to 150°C using heat lamps and pumped simultaneously to drive off adsorbed gases. Gases were extracted from the sample in a stepwise heating sequence, and noble gases and nitrogen were analysed in each temperature fraction by standard procedures<sup>14</sup>. Initial extractions (400, 600 and 800°C) were carried out in the presence of  $10^{-1}$  torr oxygen to liberate any surface-sited contaminants (400°C) and to release more labile gas components from easily combustible sites (600 and 800°C). The later steps at 1200, 1500 and 1700°C were extractions under vacuum. Blanks were run at all the extraction temperatures under identical conditions. Air standard was analysed to calibrate the mass spectrometer for sensitivity and mass discrimination.

### Results

The measured isotopic compositions of nitrogen and neon are presented in Table 2 for various temperature steps. The results for xenon isotopic composition are

**Table 2.** Nitrogen and neon isotopic compositions in Murchison microdiamond from stepwise heating mass spectrometric analysis\*

Temp. (°C)	N (ppm)	$\delta^{15}\text{N}$ (‰)	$^{22}\text{Ne}$ ( $10^{-8}$ CCSTP/g)	$^{20}\text{Ne}/^{22}\text{Ne}$	$^{21}\text{Ne}/^{22}\text{Ne}$
400 (C) <sup>†</sup>	0.84	-163.8 ± 5.6	3.24	8.791 .200	0.0491 .0044
600 (C)	56.1	-271.4 1.9	56.9	8.548 .033	0.0258 .0006
800 (C)	761	-285.0 1.9	137.4	8.375 .040	0.0321 .0008
1200 (P) <sup>‡</sup>	7399	-310.4 1.0	470.6	7.767 .034	0.0315 .0006
1500 (P)	22.2	-596.4 5.4	78.7	5.342 .027	0.0269 .0009
1700 (P)	2.3	-595.5 2.3	11.0	3.161 .020	0.0155 .0010
Total	8241	-308.6 1.1	757.8	7.622 .035	0.0306 .0007
S.W. <sup>1</sup>	—	—	—	13.6 .3	0.032 .004
Ne-A2 <sup>2</sup>	—	—	—	8.46 .12	0.0348 .0012
Ne-E <sup>2</sup>	—	—	—	< 0.1	< 0.003

\*Error in isotopic ratio is  $\pm 2\sigma$  and in concentration  $\pm 15\%$ .<sup>†</sup>C = combustion in  $10^{-1}$  torr  $\text{O}_2$ .<sup>‡</sup>P = pyrolysis.

S.W. = Solar wind

<sup>1</sup>Eberhardt *et al.*<sup>30</sup><sup>2</sup>Tang and Anders<sup>31</sup>.**Table 3.** Xenon isotopic composition in Murchison microdiamonds

Temp (°C)	$^{132}\text{Xe}$ ( $10^{-10}$ CCSTP/g)	$^{124}\text{Xe}$	$^{126}\text{Xe}$	$^{128}\text{Xe}$	$^{129}\text{Xe}$	$^{130}\text{Xe}$	$^{131}\text{Xe}$	$^{134}\text{Xe}$	$^{136}\text{Xe}$
		$^{132}\text{Xe} = 100$							
400 (C) <sup>†</sup>	19.7	0.4666 ± 0.0093	0.3747 .0075	7.792 .078	103.6 .5	15.904 .159	82.73 .41	37.591 .376	32.158 .321
600 (C)	1055	0.4534 .0036	0.4122 .0047	7.967 .032	104.3 .1	15.726 .031	82.20 .19	38.300 .151	31.830 .127
800 (C)	599	0.5739 .0191	0.4879 .0030	8.118 .033	103.7 .3	15.898 .042	82.51 .22	44.505 .175	41.000 .203
1200 (P) <sup>‡</sup>	647	0.7536 .0143	0.5164 .0104	9.040 .056	102.1 .4	16.449 .108	81.84 .30	55.529 .163	57.856 .247
1500 (P)	474	0.6254 .0097	0.5353 .0120	9.053 .040	102.6 .2	16.470 .088	81.73 .08	52.278 .100	54.227 .207
1700 (P)	52.8	0.5503 .0125	0.5230 .0760	9.107 .064	98.61 .28	17.615 .105	79.85 .93	51.594 .578	55.093 .388
Total	2848	0.5774 .0105	0.4740 .0070	8.443 .041	103.3 .2	16.085 .062	82.06 .22	46.085 .160	43.831 .190
Xe-Solar <sup>1</sup>		0.4784 .0115	0.4272 .0148	8.311 .046	104.8 .3	16.496 .043	82.28 .18	36.935 .132	29.990 .099
Xe-P <sup>2</sup>		0.464 .005	0.412 .005	8.090 .030	106.5 .3	16.00 .05	82.50 .20	37.80 .10	31.30 .10
Xe-HL <sup>3</sup>		0.774 .008	0.545 .006	9.02 .06	105.0 1.0	15.60 .03	83.90 .30	58.90 .60	64.20 .70
Xe-S <sup>2</sup>		0.069 .015	0.029 .044	21.28 .29	5.86 1.08	49.26 .54	15.37 .79	0	0

\*Error in isotopic ratio is  $\pm 2\sigma$  and in concentration  $\pm 15\%$ .<sup>†</sup>C = combustion in  $10^{-1}$  torr  $\text{O}_2$ .<sup>‡</sup>P = pyrolysis.<sup>1</sup>Eberhardt *et al.*<sup>30</sup><sup>2</sup>Tang and Anders<sup>31</sup>.<sup>3</sup>Huss<sup>32</sup>.

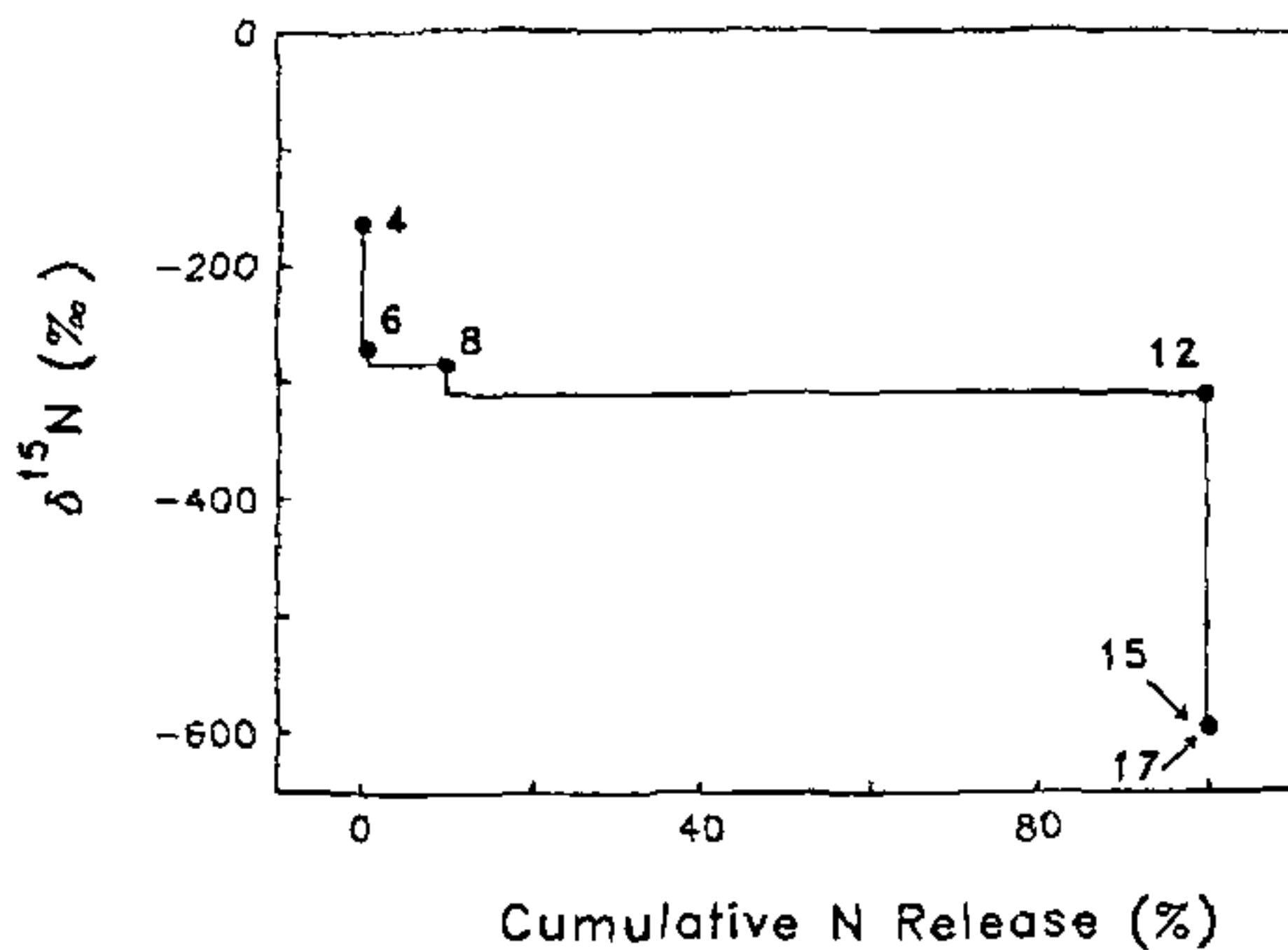


Figure 1. Temperature release pattern of nitrogen isotopes. Numbers adjacent to the data points refer to temperature in hundreds of degree centigrade.

given in Table 3. We use the  $\delta$ -notation, which is a measure of the deviation of the measured isotopic ratios in the sample from reference values, expressed in per mil (parts per thousand) unit. For example,

$$\delta^{15}\text{N} = \left[ \frac{(^{15}\text{N}/^{14}\text{N})_{\text{measured}}}{(^{15}\text{N}/^{14}\text{N})_{\text{standard}}} - 1 \right] \times 1000 \text{ (‰) (per mil)},$$

where  $(^{15}\text{N}/^{14}\text{N})_{\text{standard}}$  corresponds to that of atmospheric nitrogen and is equal to 0.0036.

### Nitrogen

The results obtained for nitrogen isotopic composition are shown in Figure 1, where we plot  $\delta^{15}\text{N}$  as a function of percentage nitrogen released at each temperature step. The data suggest the presence of two anomalous nitrogen isotopic components. The prominent component (~90% of total N) is released at the 1200°C step and has a  $\delta^{15}\text{N}$  value of -310‰. A minor second component (~0.3% of N) is released at >1200°C, where the measured  $\delta^{15}\text{N}$  value drops to -600‰.

### Noble gases

The noble gas data for isotopic compositions of neon and xenon are given in Tables 2 and 3 and are plotted in conventional three-isotope diagrams in Figures 2 and 3. Several well-defined noble gas components (Ne-E, Ne-A2, Xe-P, Xe-HL and Xe-S) are also shown in these figures for identifying specific trends in the noble gas data. The isotopic compositions of Ne-A2 and Xe-P correspond to the average composition of these elements in the solar system, the so-called planetary component, as inferred from meteorite studies. The

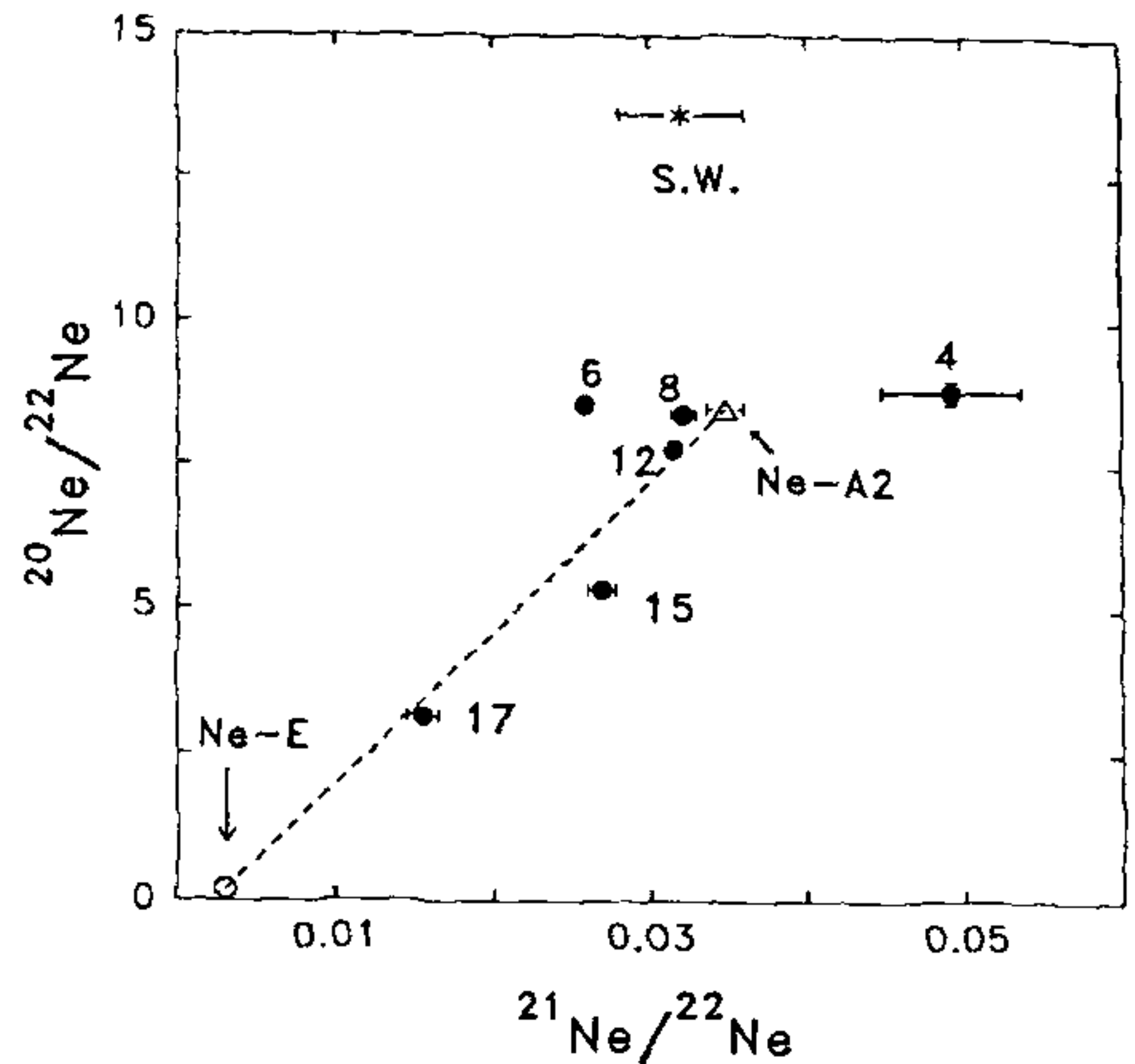


Figure 2. Three-isotope plot for the neon data. Solar wind (S.W.), Ne-A2 and Ne-E compositions are also shown. The dotted line joins the Ne-A2 and Ne-E end members.

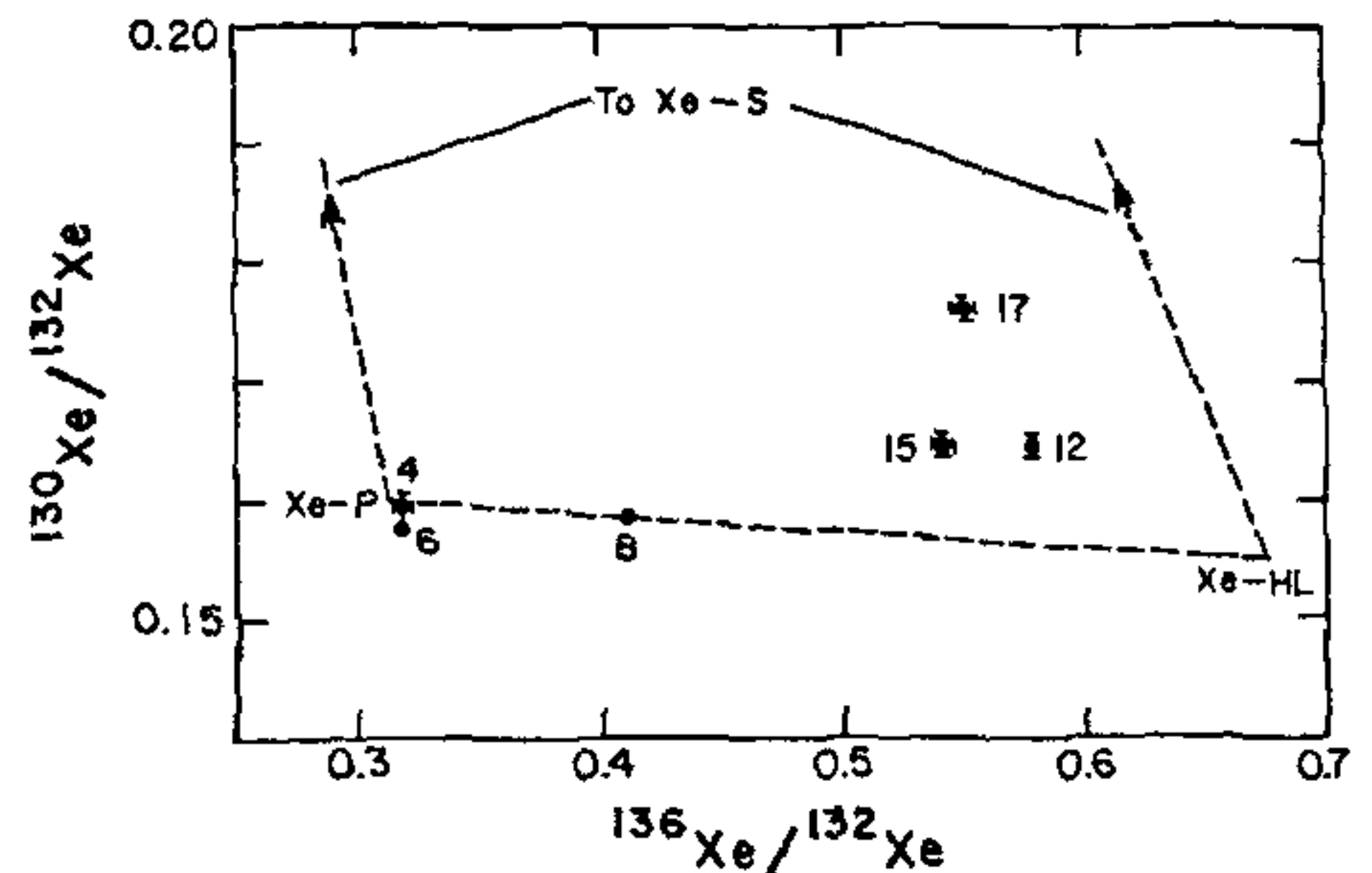


Figure 3. Three-isotope plot for the xenon data. The compositions of Xe-P and Xe-HL are also shown. Xe-S composition lies far outside the plot in the direction shown.

isotopic compositions of the other components differ significantly from the average solar composition and the labels highlight these differences, e.g. Ne-E is a component of neon with extreme isotopic composition, highly enriched in  $^{22}\text{Ne}$ , with  $^{20}\text{Ne}$  and  $^{21}\text{Ne}$  constituting <10% of the total neon in this component. Xe-HL is a xenon component that is enriched in both the heavy (H:  $^{134}\text{Xe}$ ,  $^{136}\text{Xe}$ ) and light (L:  $^{124}\text{Xe}$ ,  $^{126}\text{Xe}$ ) isotopes of xenon. The origin of this component can be traced back to stellar environments where p-process (proton capture) or photonuclear ( $\gamma, n$ ) reaction and r-process (rapid neutron capture) nucleosyntheses are operative.

Xe-S on the other hand is a component enriched in the isotopes  $^{128}\text{Xe}$  and  $^{130}\text{Xe}$  that are produced in s-process (slow neutron capture) nucleosynthesis. The specific compositions of these components are also given in Tables 2 and 3.

### Neon

The neon results are plotted in the conventional three-isotope diagram in Figure 2. The data points fall along a mixing line between the two neon components, the planetary neon (Ne-A2:  $20/22 = 8.46 \pm 0.12$ ;  $21/22 = 0.0348 \pm 0.0012$ ) and Ne-E ( $20/22 < 0.1$ ;  $21/22 = 0.003$ ). The  $400^\circ\text{C}$  point deviates from this line, suggesting an additional  $^{21}\text{Ne}$  component which most likely represents cosmogenic  $^{21}\text{Ne}$ . The low-temperature release of this component is probably due to the fact that it was recoil-implanted on the diamond surface from the adjoining silicates, where it was produced during the cosmic-ray exposure of the Murchison meteorite in interplanetary space.

### Xenon

The xenon release shows a bimodal pattern with peaks at  $600$  and  $1200^\circ\text{C}$ . Isotopic compositions of various temperature fractions show discernable differences. The shifts are very obvious for the light isotopes ( $^{124}, ^{126}\text{Xe}$ ) as well as for the heavy isotopes ( $^{134}, ^{136}\text{Xe}$ ). Both the  $400$  and  $600^\circ\text{C}$  fractions have compositions that match planetary-type xenon (Xe-P). The  $1200^\circ\text{C}$  fraction shows enrichment in both the light and the heavy isotopes of xenon (Xe-HL). The  $800^\circ\text{C}$  fraction is a mixture of Xe-P and Xe-HL. Above  $1200^\circ\text{C}$ , the Xe isotopic composition shows a shift due to the presence of yet another anomalous component (Xe-S) enriched in s-process isotopes of xenon ( $^{128}\text{Xe}$ ,  $^{130}\text{Xe}$ ). The above trends can be seen clearly in Figure 3. Note that all the data points fall in the triangle enclosed by the three xenon components: Xe-P, Xe-HL and Xe-S. In Figure 4 we show the deviation in the measured xenon isotopic ratios (normalized with respect to  $^{130}\text{Xe}$ ) from solar composition for the  $1200^\circ\text{C}$  temperature step. The presence of Xe-HL, with enrichment in both light and heavy isotopes of xenon, can be clearly seen. The xenon data for the  $800$ ,  $1500$  and  $1700^\circ\text{C}$  steps also show the presence of Xe-HL signature, although the magnitude is much lower.

### Discussion

#### *Anomalous isotopic composition of Murchison microdiamonds*

The isotopic data for nitrogen, neon and xenon for the Murchison microdiamonds show two distinctly ano-

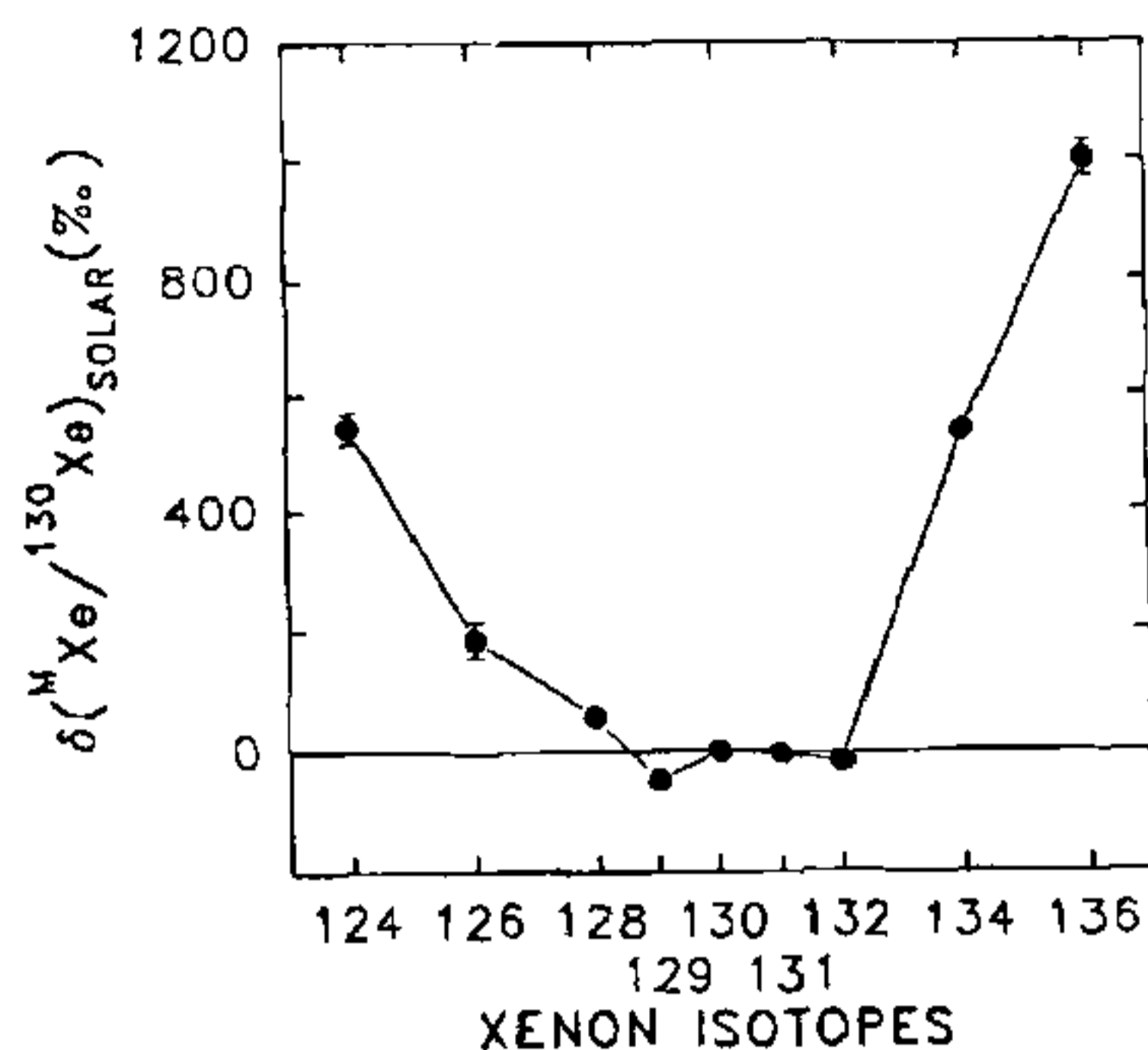


Figure 4. The deviation of xenon isotope composition from solar values (normalized with respect to  $^{130}\text{Xe}$ ) for the  $1200^\circ\text{C}$  fraction. The distinct enrichment in the lower- and higher-mass xenon isotopes (Xe-HL) can be clearly seen.

malous isotopic components at different release temperatures. The main component accounting for  $\sim 90\%$  nitrogen with a  $\delta^{15}\text{N}$  value of  $-310\text{‰}$  is seen at the  $1200^\circ\text{C}$  step and the xenon isotopic composition of this component has the distinctive Xe-HL signature. The second component is a minor fraction of the total and is released at higher temperatures ( $1500$  and  $1700^\circ\text{C}$ ). It has a much lower  $\delta^{15}\text{N}$  value of  $-600\text{‰}$  and is accompanied by Xe-S and Ne-E components. The isotopic signatures at the  $1200^\circ\text{C}$  step correspond to those observed for meteoritic microdiamonds in earlier studies<sup>1, 10-12</sup>, and confirm the interstellar origin of the Murchison microdiamonds separated by us. The Ne-E and Xe-S signals seen at high temperatures (close to the SiC release temperature) match those measured for SiC separates<sup>18</sup> and suggest the presence of a small amount of fine-grained SiC in our diamond separate. Such an admixture is generally expected in the separation procedure adopted by us, as has been noted in earlier studies<sup>19</sup>. The release of a major fraction of anomalous nitrogen as well as Xe-HL in a single temperature step ( $1200^\circ\text{C}$ ) also provides the first direct evidence for the association between the Xe-HL signature and a  $\delta^{15}\text{N}$  value of about  $-300\text{‰}$  in interstellar microdiamonds. Such an association was conjectured earlier on the basis of isotopic data for nitrogen and noble gases obtained in separate experiments<sup>17</sup>. The simultaneous measurement of nitrogen and noble gases in the present study has clearly established this association and confirmed microdiamonds to be the host for both the observed Xe-HL and nitrogen isotopic anomalies.

*Origin of interstellar diamonds: isotopic constraints*

Saslaw and Gaustad<sup>20</sup> were the first to suggest the presence of diamond in interstellar space. Thermodynamic considerations, however, predict graphite rather than diamond to be the more stable phase of carbon in interstellar environment<sup>21</sup>. Thus, one expects a higher abundance of graphite than diamond in interstellar dust. This is also consistent with the absence of direct evidence for the presence of diamond in interstellar space from astronomical observations. Contrary to such expectations, studies of primitive meteorites suggest diamond to be the most abundant component of interstellar dust preserved in meteorites. Although possibilities like destruction of graphite during early solar system processes, sampling of a nonrepresentative interstellar dust component by the solar nebula, etc., cannot be ruled out, it is interesting to note that a recent astronomical observation<sup>22</sup> suggests ubiquitous presence of diamond-like material in dense molecular clouds. This inference is based on the observation of a new infrared C-H stretching band in dense molecular clouds. This band is not seen in the diffuse interstellar medium.

The exact mode and site of formation of interstellar diamond is still uncertain, even though several plausible stellar sites have been suggested. The proposed stellar environments are carbon-rich and should also provide the anomalous isotopic signatures seen in the diamond, particularly the anomalous nitrogen isotopic composition, as nitrogen can be considered to be cogenetic with diamond. The xenon isotopic anomaly (Xe-HL) in diamond is suggestive of an imprint from supernova nucleosynthesis that can enrich the light and heavy xenon isotopes through proton capture or photoneuclear ( $\gamma, n$ ) reactions and rapid neutron capture processes, respectively. The planetary-type components (Xe-P and Ne-A2), seen at low-temperature releases, are similar to the trapped neon and xenon components found in meteorites and perhaps represent an average mix of several nucleosynthetic components. The same is probably true for the carbon isotopic composition (see Table 1), which is close to the solar system value. The initial suggestions for the source and origin of interstellar microdiamonds found in primitive meteorites attempted to couple the above findings in a plausible manner. In one scenario<sup>23</sup> it was envisaged that the formation of the diamond took place in the expanding carbon-rich He shell during the pre-supernova stage of evolution of a stellar object and the Xe-HL signature is due to chemically nonselective ion implantation on microdiamond by the subsequent supernova nucleosynthesis products. Another suggestion<sup>24</sup> involved binary star (1–2  $M_{\odot}$ ) envelopes and particular nucleosynthetic processes that can provide

the anomalous isotopic signatures. However, if diamond is indeed the most abundant component of interstellar dust and if the recent report for the ubiquitous presence of interstellar diamond in dense molecular clouds<sup>22</sup> is confirmed, one cannot have very specific settings for diamond formation, like the ones discussed above. In what follows, we propose a scenario to explain the isotopic records in diamond by assuming diamond to be a major component of interstellar dust that is concentrated particularly in dense molecular clouds.

The isotopic signatures seen in the meteoritic microdiamonds make it clear that we need a two-component model for explaining the observed isotopic anomalies in nitrogen and xenon. However, it should be emphasized that while each diamond contains about ten nitrogen atoms, the number of xenon atoms is approximately one in a million diamonds<sup>23</sup>. Thus, the xenon anomaly (Xe-HL) was most probably imprinted only in a small fraction of the diamond at a time much later than its formation. The nitrogen isotopic signal must be a characteristic feature of the formation site of the microdiamonds. It is interesting to note that the  $^{14}\text{N}/^{15}\text{N}$  values for different types of stellar environments vary over a wide scale<sup>25, 26</sup>, and this value is  $\geq 450$  for the local interstellar matter, which is much higher than the atmospheric ( $\equiv$  solar system) value of 272. We, therefore, propose that formation of diamond took place in interstellar environment that has nitrogen isotopic composition similar to that of local interstellar matter and not necessarily at very specific stellar sites. These diamonds have an anomalous nitrogen isotopic signature ( $\delta^{15}\text{N}$  of about  $-300\%$ ) and also planetary-type Xenon (Xe-P) and neon (Ne-A2) components. The Xe-HL component was implanted into a very small fraction of the diamonds at a later time during their interstellar history by plausible supernova event(s). The size of the diamond and the energy of implantation are such that the implanted component (xenon) is distributed throughout the diamond volume and gets released along with the anomalous nitrogen component. The small size (10–30 Å) of microdiamond masks effectively the possible differences in the release of nitrogen and xenon due to differences in their diffusion coefficient. The co-release of  $\sim 90\%$  of anomalous nitrogen and xenon in the 1200°C fraction is a clear indication that both of them are located in a single phase or phases that cannot be distinguished with the resolution of our stepped pyrolysis experiment. The planetary components could be of either trapped or adsorbed origin and get released at somewhat lower temperatures. Although the above scenario is compatible with the ubiquitous presence of diamond in dense molecular clouds<sup>22</sup> and can explain the observed isotopic anomalies, we still do not have a specific mechanism for the formation of diamond in interstellar

environment. Several possibilities like grain-grain collision, low-temperature chemical vapour deposition, direct condensation<sup>15, 27, 28</sup> have been proposed. However, none of them could be considered satisfactory at present. Obviously, more work, both on experimental and analytical front, will be needed for a proper understanding of the source, origin and evolution of interstellar diamonds found in primitive meteorites

## Conclusions

Microdiamonds isolated from the Murchison meteorite have anomalous nitrogen and xenon isotopic compositions attesting to their interstellar origin. The close association between the anomalous nitrogen and xenon isotopic components in interstellar diamond is evident from the co-release of these components during stepwise heating. The overabundance of interstellar diamonds in primitive meteorites compared to graphite is unexpected as graphite rather than diamond is considered to be a more stable phase of carbon in interstellar environment. However, there are recent astronomical observations suggesting ubiquitous presence of diamond-like material in dense molecular clouds. We suggest that microdiamonds found in primitive meteorites could have formed in interstellar environments with properties similar to local interstellar matter and could be a dominant component of interstellar dust. Diamond formed in such an environment will have the observed nitrogen isotopic anomaly, with depletion in <sup>15</sup>N and the associated planetary Xe and Ne components. Exposure of a small fraction of these diamonds to nucleosynthetic products of supernova, during their interstellar history prior to becoming a part of the nascent solar nebula, can explain the anomalous xenon isotopic signature in them. The exact mechanism of formation of diamond in interstellar space is yet to be understood.

1. Lewis, R. S., Tang, M., Wacker, J. F., Anders, E. and Steel, E., *Nature*, 1987, **326**, 160-162.
2. Bernatowicz, T. J., Fraundorf, G., Tang, M., Anders, E., Wopenka, B., Zinner, E. and Fraundorf, P., *Nature*, 1987, **330**, 728-730.
3. Amari, S., Anders, E., Virag, A. and Zinner, E., *Nature*, 1990, **345**, 238-240.
4. Zinner, E., Tang, M. and Anders, E., *Nature*, 1987, **330**, 730-732

5. Nichols, R. H. Jr., Hohenberg, C. M., Amari, S. and Lewis, R. S., *Meteoritics*, 1991, **26**, 377-378.
6. Bernatowicz, T. J., Gibbons, P. C. and Lewis, R. S., *Astrophys J.*, 1990, **359**, 246-255.
7. Gallino, R., Busso, M., Picchio, G. and Raiteri, C. M., *Nature*, 1990, **348**, 298-302.
8. Brown, L. E. and Clayton, D. D., *Astrophys. J.*, 1992, **392**, L79-L82
9. Tang, M., Anders, E., Hoppe, P. and Zinner, E., *Nature*, 1989, **339**, 351-354.
10. Alexander, C. M. O'D., Arden, J. W., Ash, R. D. and Pillinger, C. T., *Earth Planet. Sci. Lett.*, 1990, **99**, 220-229.
11. Tang, M. and Anders, E., *Geochim. Cosmochim. Acta*, 1988, **52**, 1235-1244
12. Schelhaas, N., Ott, U. and Begetmann, F., *Geochim Cosmochim Acta*, 1990, **54**, 2869-2882.
13. Nichols, R. H. Jr., Hohenberg, C. M., Alexander, C. M. O'D., Olinger, C. T. and Arden, J. W., *Geochim. Cosmochim. Acta*, 1991, **55**, 2921-2936.
14. Murty, S. V. S. and Goswami, J. N., *Proc. Lunar Planet. Sci. Conf.*, 1992, **22**, 225-237.
15. Murty, S. V. S., PRL Technical Note (Physical Research Laboratory, Ahmedabad) TN-88-59, 1988, 36 pp
16. Lattimer, J. M., Schramm, D. N. and Grossman, L., *Astrophys. J.*, 1978, **219**, 230-249.
17. Tang, M., Lewis, R. S., Anders, E., Grady, M. M., Wright, I. P. and Pillinger, C. T., *Geochim Cosmochim. Acta*, 1988, **52**, 1221-1234
18. Zinner, E., Tang, M. and Anders, E., *Geochim. Cosmochim Acta*, 1989, **53**, 3273-3290.
19. Lewis, R. S., Anders, E. and Bruce, T. B., *Nature*, 1989, **339**, 117-121.
20. Saslaw, W. C. and Gaustad, J. E., *Nature*, 1969, **221**, 160-162.
21. Bundy, F. P., Strong, H. M. and Wentorf, R. H., in *Chemistry and Physics of Carbon* (ed. Throwen, P. A.), Dekker, New York, 1973, vol 18, p 1.
22. Allamandola, L. J., Sandford, S. A., Tielens, A. G. G. M. and Herbst, T. M., *Science*, 1993, **260**, 64-66.
23. Clayton, D. D., *Astrophys J.*, 1989, **340**, 613-619.
24. Jorgensen, U. G., *Nature*, 1988, **332**, 702-705.
25. Gusten, R. and Ungerechts, H., *Astron. Astrophys.*, 1985, **145**, 241-250
26. Wannier, P. G., Anderson, B. G., Olofsson, H., Ukita, N. and Young, K., *Astrophys. J.*, 1991, **380**, 593-605.
27. Tielens, A. G. G. M., Seab, C. G., Hollenbach, D. J. and McKee, C. F., *Astrophys. J.*, 1987, **319**, L109-L113.
28. Angus, J. C. and Haymann, C. C., *Science*, 1988, **241**, 913-921.
29. Anders, E., in *Meteorites and the Early Solar System* (eds. Kerridge, J. F. and Mathews, M. S.), University of Arizona Press, Arizona, 1988, pp 927-955.
30. Eberhardt, P., Geiss, J., Graf, H., Grogler, N., Mendia, M. D., Morgeli, M., Schwaller, H., Stettler, A., Krahenbuhl, U. and Von Gunter, H. R., *Proc. Lunar Sci. Conf.*, 1972, **3**, 1821-1856.
31. Tang, M. and Anders, E., *Geochim. Cosmochim. Acta*, 1988, **52**, 1245-1254.
32. Huss, G., *Nature*, 1990, **347**, 159-162.

Received 28 June 1993; revised accepted 5 October 1993