An enhanced cosmic-ray flux towards ζ Persei inferred from a laboratory study of the H_3^+–e^- recombination rate

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The H_3^+ molecular ion plays a fundamental role in interstellar chemistry, as it initiates a network of chemical reactions that produce many molecules[1,2]. In dense interstellar clouds, the H_3^+ abundance is understood using a simple chemical model, from which observations of H_3^+ yield valuable estimates of cloud path length, density and temperature[3,4]. But observations of diffuse clouds have suggested that H_3^+ is considerably more abundant than expected from the chemical models[5–7]. Models of diffuse clouds have, however, been hampered by the uncertain values of three key parameters: the rate of H_3^+ destruction by electrons (e^-), the electron fraction, and the cosmic-ray ionization rate. Here we report a direct experimental measurement of the H_3^+ destruction rate under nearly interstellar conditions. We also report the observation of H_3^+ in a diffuse cloud (towards ζ Persei) where the electron fraction is already known. From these, we find that the cosmic-ray ionization rate along this line of sight is 40 times faster than previously assumed. If such a high cosmic-ray flux is ubiquitous in diffuse clouds, the discrepancy between chemical models and the previous observations[5–7] of H_3^+ can be resolved.

The dissociative recombination of H_3^+ (that is, the exothermic reaction $H_3^+ + e^- \rightarrow H + H + H$ or $H_2 + H$) has been one of the most controversial topics in the field of ion physics[8,9], as different experimental methods have disagreed by a factor of 10,000 on the value of the thermal rate coefficient. In addition, it was only in the past few years that a plausible theoretical mechanism (the "indirect process"[10]) was used to explain the dissociative recombination of H_3^+ in its lowest vibrational level[11]. However, those calculations gave a cross-section[12] 1,000 times smaller than those measured by the ion storage rings CRYRING[13] (Sweden), ASTRID[14] (Denmark) and TARN II[15] (Japan), which all gave essentially the same result. Calculations[16] accounting for the symmetry-breaking of the equilateral-triangle-shaped H_3^+ ion by the incoming electron were in better accord with the experiments, and a more detailed theoretical treatment[17] has now provided a step forward in terms of agreement, although important discrepancies with experiment remain to be resolved.

Meanwhile, a series of experiments at the Test Storage Ring in Germany[18,19] and at CRYRING[20] suggested that, whereas H_3^+ undergoes complete vibrational relaxation by spontaneous emission of radiation in storage-ring experiments, the high rotational temperature of H_3^+ affected the measured values of the dissociative recombination rate coefficient in the earlier experiments[13–15]. These experiments[18–20] thus made clear that a measurement of a thermal rate coefficient applicable to interstellar clouds would have to be performed on rotationally cold H_3^+. This represented a challenge to the ion storage ring technique, which has traditionally used hot-filament or hollow-cathode ion sources that produce ions at much higher rotational temperatures than those of diffuse clouds.

The challenge of producing rotationally cold H_3^+ ions has been addressed by the development of a supersonic expansion ion source (see Fig. 1 top trace for results). The measured rate coefficient for the dissociative recombination of H_3^+ using this source is shown in Fig. 2. The general shape is similar to that reported in earlier studies[13–15], but the present data show a number of resonances between 1 meV and 1 eV. Although hints of resonance structures have been reported previously[20], the present experiment is, to our knowledge, the first to use rotationally cold ions, which apparently enhance the structure. Figure 3 shows the derived rate coefficient for a thermal electron distribution (k_e), which is about 40% lower than that measured in storage-ring studies of rotationally hot H_3^+ ions[13,14].

Whereas different ion storage ring experiments have consistently yielded about the same value of $k_e \approx 1 \times 10^{-7}\ \mathrm{cm^3\,s^{-1}}$ at 300 K (apart from the present experiment, which uses rotationally cold ions), afterglow experiments, which indirectly infer k_e from the removal of electrons in a decaying plasma of hydrogen and noble gases, are more difficult to interpret. Different afterglow measurements[21] since 1984 have yielded values of k_e (at 300 K) which range from $<10^{-11}$ to $\sim 2 \times 10^{-7}\ \mathrm{cm^3\,s^{-1}}$. The reason for this wide

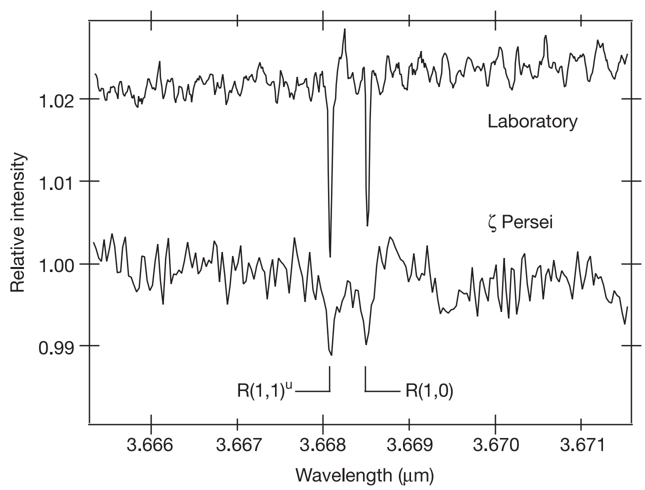

Figure 1 Spectra of two H_3^+ transitions arising from the two lowest rotational levels, which are the only levels with significant population in diffuse clouds. $R(1,1)^u$ originates from the lowest *para* level ($J = 1$, $K = 1$), while R(1,0) comes from the lowest *ortho* level ($J = 1$, $K = 0$). Note that the ($J = K = 0$) level is forbidden by the Pauli principle. For energy-level diagrams and a description of the H_3^+ notation, see ref. 28. Top trace, cavity ringdown[29] laser absorption spectrum of the supersonic expansion ion source. In this source, H_3^+ number densities of $\sim 10^{11}\ \mathrm{cm^{-3}}$ were produced in a direct-current discharge plasma downstream of a 500-μm pinhole through which hydrogen gas (at 2.5 atm) expanded supersonically into a vacuum. Depending on conditions, the rotational temperature of the plasma was 20–60 K, on the basis of the relative intensities of the two transitions. This laboratory spectrum has been smoothed and multiplied by a factor of 1,000 for clarity. Bottom trace, spectrum of a diffuse cloud towards ζ Persei obtained using the CGS4 infrared spectrometer at the United Kingdom Infrared Telescope (UKIRT) on the night of 2001 September 5 UT using our standard observing techniques[7]. The total on-source integration time was 40 min, and the ζ Persei spectrum has been divided by that of a standard star (BS936) to remove absorption lines due to the Earth's atmosphere. The spectrum is displayed in rest wavelengths, and indicates an H_3^+ column density (the integral of the number density along the line of sight) of $N(H_3^+) = 8 \times 10^{13}\ \mathrm{cm^{-2}}$. The ratio of the two absorption lines yields a temperature estimate of 23 K.

variance in afterglow measurements is still not understood, although it may be related to the complicated modelling and analysis that goes into extracting the value of k_e from the experimental measurements. In contrast, the storage-ring experiments are conceptually very simple, and require a comparatively straightforward analysis to determine k_e, so we consider the present experiment to provide a much more robust measurement of the appropriate value of k_e for interstellar conditions. It is important to note two differences between the present experiment and interstellar conditions: the presence of high electron number densities ($n(e^-) \approx 10^7\,cm^{-3}$) and magnetic fields ($\sim$300 G). Because these are inherent to the storage-ring technique and cannot be avoided, our results need to be confirmed by theoretical calculations. Although there are still some unexplained differences between theory and our experiment, the most recent calculations[17] yield a value of k_e ($\sim 2\times10^{-7}\,cm^3\,s^{-1}$ at 40 K) that is similar to our results. Although we cannot explain the wide range in afterglow results, the weight of the evidence suggests that we have determined the appropriate value of k_e for interstellar conditions, and this fact compels us to examine the implications of adopting this new value for modelling H_3^+ chemistry in diffuse clouds.

The chemical model for H_3^+ in diffuse clouds is quite simple, in contrast to the involved models needed for most other molecules. H_3^+ is formed when H_2 is ionized by a cosmic ray (a high-energy proton or electron) to form H_2^+, which then undergoes a very rapid ion–neutral reaction with another H_2 to form H_3^+ and an H atom.

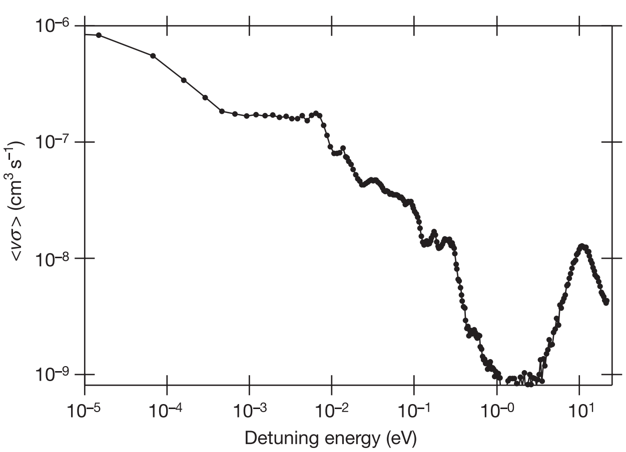

Figure 2 Measured dissociative recombination rate coefficient of rotationally cold H_3^+ as a function of detuning energy. For these measurements, the supersonic expansion ion source (described in Fig. 1 legend) was mounted onto the MINIS ion injection endstation of the CRYRING ion storage ring at the Manne Siegbahn Laboratory in Stockholm. The ions leaving the source were mass-selected and accelerated to 300 keV per a.m.u. before being injected into the ring. After injection, the ions were accelerated to their final energy (12.1 MeV), which gave a circulating ion beam current of 0.31 μA. In one straight section ('interaction region') of the 52-m-circumference storage ring, the ions passed collinearly through an approximately homogeneous electron beam of 85 cm length and 4 cm diameter ('electron cooler'). The electron beam current was 35.5 mA and the electron energy (2.2 keV) was chosen so that ion–electron velocity matching was achieved. For the first five seconds after the ion beam had reached its full energy, the electron beam acted to reduce the phase-space volume of the stored ion beam, leading to a narrow ion-velocity spread and a correspondingly small ion beam diameter ($\sim$1 mm). During this time, some stored ions were lost to dissociative recombination with electrons and collisions with residual gas molecules, while the remaining ions relaxed to the ground vibrational state by spontaneous emission. This cooling phase was followed by a measurement of the total number of neutral reaction products (H + H + H and H_2 + H) using a surface barrier detector mounted tangentially to the ring following the interaction region. The electron energy was varied by changing the cathode voltage in the electron cooler, and the neutral fragment signal was recorded as a function of the longitudinal energy difference between electrons and ions ('detuning energy'). Note the resonance structure between 1 meV and 1 eV that was only hinted at in previous experiments using rotationally hot ions.

The rate of formation can be expressed as $\zeta n(H_2)$, where ζ is the cosmic-ray ionization rate and $n(H_2)$ is the number density of H_2. The destruction of H_3^+ in diffuse clouds is dominated by dissociative recombination, and the rate of this process can be expressed as $k_e n(e^-) n(H_3^+)$. In steady state[7], the H_3^+ number density can then be written as $n(H_3^+) = (\zeta/k_e) n(H_2)/n(e^-)$.

Astronomical absorption spectra, however, yield direct information only about the absorber's column density (the integral of the number density along the line of sight), not the number density itself. We assume that the ratio $n(H_2)/n(e^-)$ is approximately constant throughout the cloud, and replace it by the ratio of column densities $N(H_2)/N(e^-)$. Given this assumption, $n(H_3^+)$ is itself a constant, and we write $N(H_3^+) = n(H_3^+)L$, where L is the absorption path length. Substitution into the above equation then yields the relation $\zeta L = k_e[N(e^-)/N(H_2)]N(H_3^+)$.

Here we report the observation of H_3^+ in the diffuse cloud towards ζ Persei (Fig. 1 bottom trace); this is, to our knowledge, the first detection of H_3^+ along a line of sight where the electron fraction can be estimated. Assuming that most electrons come from the photoionization of carbon, we can use existing ultraviolet measurements of molecular hydrogen[22] and ionized carbon[23] to derive the electron fraction $N(e^-)/N(H_2) = 3.8\times10^{-4}$. Combining this electron fraction, our observed H_3^+ column density, and the value $k_e = 2.6\times10^{-7}\,cm^3\,s^{-1}$ from the CRYRING experiment (Fig. 3), we find $\zeta L \approx 8{,}000\,cm\,s^{-1}$ along this line of sight.

There is little direct observational information about the value of either ζ or L. It has generally been assumed that the value of ζ in diffuse clouds is the same as in dense clouds, where various measurements suggest a value of $\zeta = 3\times10^{-17}\,s^{-1}$. The absorption path length can be indirectly estimated by dividing the total column density of hydrogen nuclei by an assumed number density. The total column density towards ζ Persei is $N_H \equiv N(H) + 2N(H_2) \approx 1.6\times10^{21}\,cm^{-2}$ (adopting $N(H) = 6.3\times10^{20}\,cm^{-2}$, ref. 24). The number density can be estimated from the rotational excitation of the CO and C_2 molecules, as well as from more detailed cloud models that consider the H/H_2 ratio[25]. Although each of these methods has its uncertainties, they are all consistent with an average number density of $\langle n_H \rangle = 150$–$500\,cm^{-3}$. If we adopt a density of $\langle n_H \rangle = 250\,cm^{-3}$, we obtain a path length of $L = 2.1$ pc.

Together, the 'canonical' value of ζ and the best estimate of L yield $\zeta L = 200\,cm\,s^{-1}$. However, the measured H_3^+ column density implies $\zeta L = 8{,}000\,cm\,s^{-1}$. If we adopt the canonical value of ζ,

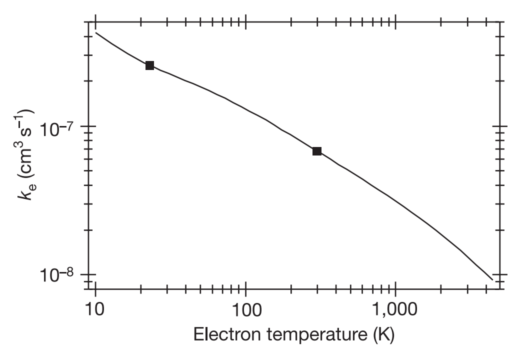

Figure 3 Calculated thermal rate coefficient k_e for the dissociative recombination of rotationally cold H_3^+ ions as a function of electron temperature, based on CRYRING measurements. The experimentally measured rate coefficient ($\langle v\sigma \rangle$) shown in Fig. 2 was deconvolved using the known electron velocity distribution, yielding the dissociative recombination cross-section σ as a function of electron energy. The cross-section was then integrated over a Boltzmann distribution of energies (corresponding to a thermalized electron energy distribution) to yield the 'thermal' rate coefficient at each value of the electron temperature. The values $k_e(23\,K) = 2.6\times10^{-7}$ and $k_e(300\,K) = 6.8\times10^{-8}\,cm^3\,s^{-1}$ are indicated with squares. Note that the value of $k_e(300\,K)$ refers to an electron temperature of 300 K, but still pertains to rotationally cold H_3^+ ions. Details of the experimental method and analysis will be presented elsewhere.

we would have to accept a very long path length of $L = 85$ pc and a low density of $\langle n_H \rangle = 6\,cm^{-3}$. On the other hand, if we adopt the canonical path length, we must increase the ionization rate to $\zeta = 1.2 \times 10^{-15}\,s^{-1}$. Given the evidence pointing to a short path length, and the fact that ζ is essentially unconstrained in diffuse clouds, we consider the higher ionization rate to be the more likely explanation. A higher value of ζ for diffuse clouds may also find some support from considerations of the abundance of the OH molecule[26]. A more detailed discussion of the ζ Persei line of sight, as well as other observations of H_3^+ in diffuse clouds, will be presented elsewhere.

The high value of ζ suggested by the CRYRING measurements and the ζ Persei observations can be reconciled with the lower ionization rate in dense clouds by postulating the existence of a large flux of low-energy cosmic rays that can penetrate diffuse clouds but not dense clouds. Such a cosmic-ray flux would bring the models and observations of H_3^+ into agreement, but would also have far-reaching implications for the chemistry and physics of interstellar clouds. From a chemical perspective, a higher value of ζ would proportionately increase the number densities of oxygen compounds such as OH, as well as affecting the relative abundances of deuterium-bearing molecules. The high number density of H_3^+ in diffuse clouds would also increase the rate of proton-transfer reactions, which play the key role in the formation of complex molecules in dense clouds, but which have been considered relatively unimportant in diffuse clouds. From a physical perspective, a higher cosmic-ray ionization rate represents an additional heating source for interstellar gas, and could have a significant impact on the thermal balance of the warm neutral medium[27]. Further observations of H_3^+, H_2 and C^+ in lines of sight to other diffuse clouds are needed to determine if the ζ Persei line of sight is unusual, or whether a large, low-energy cosmic ray flux indeed pervades our galaxy. □

Received 19 November 2002; accepted 17 February 2003; doi:10.1038/nature01498.

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Acknowledgements B.J.M. and A.J.H. thank K. Wilson and C.-Y. Chung for their assistance in the laboratory, D. Lucas for the loan of equipment, and H. Chan and E. Granlund for their support. B.J.M. thanks T. Oka, L. M. Hobbs, D. G. York and T. P. Snow for conversations about the ζ Persei line of sight. We thank C. H. Greene for providing us with the results of his calculations in advance of publication. M.L. thanks D. Zajfman for information on TSR results before publication. The idea of using a supersonic expansion source for dissociative recombination measurements originated during conversations between B.J.M. and C. M. Lindsay, and the experiment was designed by B.J.M., A.J.H. and M.L. The supersonic expansion ion source was built and spectroscopically characterized by A.J.H. and B.J.M. in the laboratory of R.J.S. in Berkeley. The dissociative recombination measurements in Stockholm were carried out by all of the authors except R.J.S. and T.R.G. The UKIRT observations were obtained by B.J.M. and T.R.G. The authors thank the staff of the Manne Siegbahn Laboratory for help with the experiment. B.J.M. is supported by the Miller Institute for Basic Research in Science. A.J.H. and R.J.S. acknowledge support from the AFOSR and NSF. T.R.G. is supported by Gemini Observatory, operated by AURA for the international Gemini partnership. N.D. and G.H. were supported in part by the US DOE, Office of Fusion Energy. J.S. is supported in part by the State Committee for Scientific Research. Support is acknowledged from the EU fifth framework program, the EOARD, the Swedish Research Council and STINT.

Competing interests statement The authors declare that they have no competing financial interests.

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Structure and thermal history of the H-chondrite parent asteroid revealed by thermochronometry

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Our Solar System formed ~4.6 billion years ago from the collapse of a dense core inside an interstellar molecular cloud. The subsequent formation of solid bodies took place rapidly. The period of <10 million years over which planetesimals were assembled can be investigated through the study of meteorites[1–3]. Although some planetesimals differentiated and formed metallic cores like the larger terrestrial planets, the parent bodies of undifferentiated chondritic meteorites experienced comparatively mild thermal metamorphism that was insufficient to separate metal from silicate[4,5]. There is debate about the nature of the heat source[6–9] as well as the structure and cooling