the presence of loosely bound neutrons as might be inferred from the sequences $^{16}$O + $^{144,154}$Sm, $^{40}$Ar + $^{58,60,62,64}$Ni, and $^{86}$Kr + $^{58,62,64}$Ni, or to the nuclear stiffness which is believed to change appreciably within the series $^{12}$C + Pd, Sn, Te, and $^{40}$Ar + Pd, Sn, Te when crossing the $Z = 50$ shell closure. However, the intrinsic nuclear structure of the colliding nuclei does not drastically offset the general increase of the near-barrier cross section. Moreover, its relative significance seems to diminish the heavier the systems. The data for $^{35}$Cl, $^{40}$Ca + $^{58,\cdots,64}$Ni and $^{40}$Ar + $^{144,148,154}$Sm (Refs. 1 and 3), if analyzed in a similar way, would not invalidate the present systematics.

In conclusion, we find that heavy-ion fusion in the near-barrier regime by far exceeds the prediction of the simple concept of a potential barrier which depends on the separation coordinate only. The excess shows a global increase for heavier systems. The reduced radius of the coalescing system, reflecting the size of the opposing surfaces, provides a good scaling[18] to this phenomenon thus indicating shape distortion or neck formation.

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# Detection of the Infrared Fundamental Band of HeH$^+$

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Nine vibration-rotation transitions of the fundamental band of HeH$^+$ have been observed in absorption by using a tunable, infrared, difference-frequency laser source. The molecular constants for the $v = 0$ and $v = 1$ levels of the $X\,^1\Sigma^+$ state have been determined and they agree very will with those derived from the *ab initio* calculation of Bishop and Cheung.

Recently Oka[1] observed the vibration-rotation spectrum of H$_3^+$ using a tunable infrared source made by mixing radiation from an argon-ion laser and a dye laser in a LiNbO$_3$ crystal.[2] It was suggested by Oka that other ions might be detected with similar methods. We would like to report the observation of nine lines, $P(4)(J = 3 \to 4)$ to $R(4)(J = 5 \to 4)$, of the fundamental band ($v = 1 \to 0$)

of HeH$^+$.

HeH$^+$ is the simplest closed-shell heteronuclear molecule and is isoelectronic with H$_2$. Because the two most abundant elements in the universe are hydrogen and helium, it is not surprising that HeH$^+$ has been subjected to intense theoretical and considerable experimental scrutiny.[3,4]
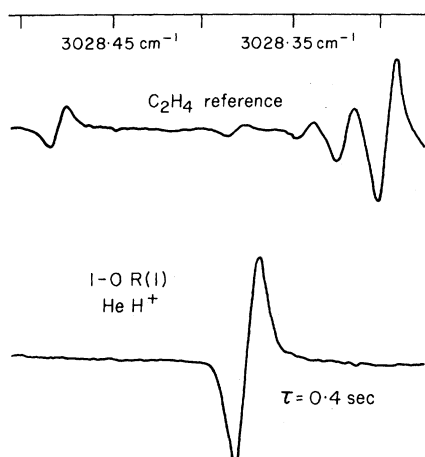
Hogness and Lunn[5] discovered HeH$^+$ in 1924

FIG. 1. The experimental trace of the $R(1)$ line of the 1-0 infrared band of HeH$^+$ at 3028.3750 cm$^{-1}$. The time constant was 0.4 sec.

with a mass spectrometer. Until the work of Tolliver, Kyrala, and Wing[3] in 1979, however, no high-resolution spectroscopic data were available. They used Doppler-tuned ion-beam spectroscopy with a CO laser to detect the $P(12)$ and $P(13)$ lines of the fundamental vibration-rotation band as well as three lines of the 2-1 band. Since then they have observed more lines in higher members of the same ($\Delta v = 1$) sequence.[6] Unfortunately, because there are no CO laser lines in the appropriate spectral region, the astrophysically important $R(0)$ and $P(1)$ lines of the fundamental band could not be observed. In fact, because good spectroscopic constants cannot be extracted from only two high-$J$ $P$-branch lines, $R(0)$ and $P(1)$ are not

accurately predicted by their experimental results.

Our experimental setup was essentially identical to that of Oka.[1] The infrared radiation was generated as described for H$_3{}^+$ with the exception that a Coherent 699-21 ring dye laser was used instead of a Spectra-Physics 580A laser. This provided an increase in infrared power (about a factor of 3) and, more importantly, an increase in the absolute accuracy with which transition frequencies can be measured (now about $\pm 0.001$–$0.002$ cm$^{-1}$). The reference gases used for frequency calibration were C$_2$H$_4$,[7] H$_2$CO,[7] and N$_2$O.[8] The infrared source was frequency modulated at 2.5 kHz with an amplitude of about 400 MHz.

We used a liquid-nitrogen–cooled multiple-reflection discharge cell[1] with a mixture of about 100:1 of He to H$_2$. The HeH$^+$ absorption signal was very sensitive to the exact mixing ratio, which was optimized experimentally. This sensitivity is presumably caused by the very rapid reaction[9]

$$\text{HeH}^+ + \text{H}_2 \rightarrow \text{He} + \text{H}_3{}^+,$$
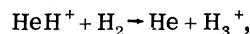
which removes the HeH$^+$ at small He-to-H$_2$ mixing ratios. HeH$^+$ is probably produced by the reactions[10]

$$\text{He}^* + \text{H}_2 \rightarrow \text{HeH}^+ + \text{H} + e$$

and[11]

$$\text{H}_2{}^+(v \gtrsim 3) + \text{He} \rightarrow \text{HeH}^+ + \text{H}.$$
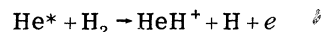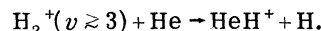
Our total pressure was 0.45 Torr and the optimum current density was about 30 mA/cm$^2$. The path length was 32 m and the maximum absorption [for

TABLE I. Observed and calculated wave numbers of the HeH$^+$ $v = 1 \leftarrow 0$ band (in inverse centimeters). The numbers in parentheses next to the observed column are the obs.–calc. differences obtained by use of the spectroscopic constants of Table II.

| Transition | Obs. | *Ab initio* calc.[a] | Obs.–Calc. |
|---|---|---|---|
| $R(4)$ | 3157.2967(−1) | 3157.632 | −0.335 |
| $R(3)$ | 3121.0765(1) | 3121.417 | −0.341 |
| $R(2)$ | 3077.9919(4) | 3078.332 | −0.340 |
| $R(1)$ | 3028.3750(−8) | 3028.715 | −0.340 |
| $R(0)$ | 2972.5732(0) | 2972.913 | −0.340 |
| $P(1)$ | 2843.9035(13) | 2844.242 | −0.339 |
| $P(2)$ | 2771.8059(−9) | 2772.140 | −0.334 |
| $P(3)$ | 2695.0500(−5) | 2695.381 | −0.331 |
| $P(4)$ | 2614.0295(4) | 2614.353 | −0.324 |
| $P(12)$[b] | 1855.905(0) | 1856.152 | −0.247 |
| $P(13)$[b] | 1751.971(0) | 1752.198 | −0.227 |

[a]From Ref. 4.      [b]From Ref. 3.

TABLE II. Observed and calculated spectroscopic constants (in inverse centimeters). 1-$\sigma$ uncertainty in the last digits is given in parentheses.

| | $v = 0$ | | $v = 1$ | |
| --- | --- | --- | --- | --- |
| | Obs. | Calc.[a] | Obs. | Calc.[a] |
| $B_v$ | 33.558 41 (21) | 33.559 161 (75) | 30.839 91 (18) | 30.840 008 (43) |
| $D_v \times 10^2$ | 1.621 03 (122) | 1.620 29 (15) | 1.586 94 (103) | 1.584 133 (84) |
| $H_v \times 10^6$ | 5.81 (15) | 5.840 (10) | 5.76 (20) | 5.373 6 (56) |
| $L_v \times 10^9$ | $\cdots$ | $-2.870$ (21) | $\cdots$ | $-3.218$ (12) |
| $T_v$ | 0.0 | $-0.000$ 83 (95) | 2910.956 81 (64) | 2911.297 03 (54) |

[a]Calculated from the *ab initio* term values of Ref. 4 for $0 \leqslant J \leqslant 15$.

$R(1)$] was about 4%. This leads to an estimate of about $10^9$ molecules/cm$^3$ for the density of HeH$^+$ using the *ab initio* dipole moment function.[12] The relative strength of the rotational lines suggests a rotational temperature of about 200 K while the linewidths are consistent with a translational temperature of about 200 K.

Figure 1 is an experimental trace of the $R(1)$ line. The nine observed transitions of the $v = 1$ $\to 0$ band of the $X^1\Sigma^+$ state of HeH$^+$ are presented in Table I together with the two transitions observed by Tolliver, Kyrala, and Wing.[3] The lines were fitted by using the power-series expression for vibration-rotation term values of $^1\Sigma$ molecules:

$$F_v(J) = T_v + B_v J(J+1) - D_v [J(J+1)]^2$$
$$+ H_v [J(J+1)]^3 + L_v [J(J+1)]^4. \qquad (1)$$

The values of these parameters are given in Table II. It was necessary to include terms up to $H$ and the standard deviation of the fit was 0.0005 cm$^{-1}$.

The *ab initio* calculated transition frequencies shown in Table I were computed from Bishop and Cheung's $v = 0$ and $v = 1$ term values.[4] They included adiabatic corrections to the Born-Oppenheimer potential in their calculation. Our transition frequencies are about 0.33 cm$^{-1}$ lower than their predictions as shown in the last column of Table I. This is excellent agreement (1 part in $10^4$) and well within their modest estimate of 1 cm$^{-1}$ accuracy.

In order to compare the constants of Table II with theoretical predictions,[4] the term values for $0 \leqslant J \leqslant 15$ for $v = 0$ and 1 were fitted to Eq. (1). The agreement for all constants, except the band origin, is within 2 or 3 standard deviations (see Table II).

One of the reasons HeH$^+$ is of interest is the possibility that it might be found in extraterrestrial sources.[12] The first allowed electronic tran-

sition $A^1\Sigma^+$-$X^1\Sigma^+$ in the vacuum ultraviolet is expected to be very difficult to observe.[12] Thus a high-resolution infrared search for $R(0)$ of the fundamental band seems more promising. There is considerable controversy about the detectability of HeH$^+$.[13] However, the recent calculations of Roberge and Dalgarno[14] suggest that HeH$^+$ can be detected in dense clouds subjected to x-ray and extreme ultraviolet radiation and in planetary nebulae.

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