LETTER TO THE EDITOR

Revised spectroscopic parameters of SH$^+$ from ALMA$^*$ and IRAM 30 m** observations

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1. Introduction

Hydrides, which consist of one heavy atom and one or more H atoms, are of fundamental importance in the interstellar medium (ISM). Unfortunately, their fundamental transitions at sub-millimeter (sub-mm) wavelengths are often difficult to observe from the ground. The Herschel Space Observatory and the Stratospheric Observatory for Infrared Astronomy (SOFIA) have increased our knowledge about hydrides in space considerably. Recent detections include ArH$^+$ with Herschel (Barlow et al. 2013) and SH with SOFIA (Neufeld et al. 2012).

The reactive molecular ion SH$^+$ (sulfanylium) is an interesting probe of energetic processes in the ISM. In particular, SH$^+$ is only detectable in significant amounts if the very high endothermicity (0.86 eV or ~9860 K) of the gas-phase reaction

\[ S^+ + H_2(v = 0) \rightarrow SH^+(X^3\Sigma^-) + H \]  

(1)

can be overcome. Using the Atacama Pathfinder EXperiment (APEX) 12 m telescope, Menten et al. (2011) discovered recently sub-mm SH$^+$ absorption lines in the envelope of the prototypical star-forming region Sagittarius B2(M), Sgr B2(M) for short, a very strong continuum source close to the Galactic center, and in the diffuse interstellar clouds on the line of sight toward this source. The ubiquity of SH$^+$ in diffuse clouds was emphasized soon thereafter by Herschel/HIFI observations toward several Galactic sight lines by Godard et al. (2012). These authors proposed that the required energy for reaction (1) arises from turbulent dissipation, shocks, or shears in these very low density clouds. Sub-mm SH$^+$ emission lines were also detected by Herschel in higher density environments such as the high-mass star-forming region W3 IRS5 (Benz et al. 2010) or the Orion Bar photo-dissociation region (PDR) (Nagy et al. 2013). These are strongly UV-irradiated environments where the gas attains high temperatures ($\lesssim$ 1000 K) and where H$_2$ molecules are UV-pumped to vibrationally excited states. In addition, in these PDR environments, reaction (1) becomes exothermic when H$_2$ molecules are in the $v = 2$ or higher vibrational levels, thus enhancing the SH$^+$ abundance (e.g., Zanchet et al. 2013). Finally,

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ABSTRACT

Hydrides represent the first steps of interstellar chemistry. Sulfanylium (SH$^+$), in particular, is a key tracer of energetic processes. We used ALMA and the IRAM 30 m telescope to search for the lowest frequency rotational lines of SH$^+$ toward the Orion Bar, the prototypical photo-dissociation region illuminated by a strong UV radiation field. On the basis of previous Herschel/HIFI observations of SH$^+$, we expected to detect emission of the two SH$^+$ hyperfine structure (HFS) components of the $N_J = 1_{0} - 0_{1}$ fine structure (FS) component near 346 GHz. While we did not observe any lines at the frequencies predicted from laboratory data, we detected two emission lines, each ~15 MHz above the SH$^+$ predictions and with relative intensities and HFS splitting expected for SH$^+$. The rest frequencies of the two newly detected lines are more compatible with the remainder of the SH$^+$ laboratory data than the single line measured in the laboratory near 346 GHz and previously attributed to SH$^+$. Therefore, we assign these new features to the two SH$^+$ HFS components of the $N_J = 1_{0} - 0_{1}$ FS component and re-determine its spectroscopic parameters, which will be useful for future observations of SH$^+$, in particular if its lowest frequency FS components are studied. Our observations demonstrate the suitability of these lines for SH$^+$ searches at frequencies easily accessible from the ground.

Key words. ISM: individual objects: Orion Bar – radio lines: ISM – ISM: molecules – molecular data – line: identification
if doubly ionized sulfur atoms co-exist with H$_2$ molecules in X-ray dissociation regions (XDRs), such as Galactic center clouds (e.g., Godard et al. 2012; Etxaluze et al. 2013), the very exothermic reaction

$$\text{S}^{++} + \text{H}_2 \rightarrow \text{SH}^+ + \text{H}^+ + 8.79 \text{ eV}$$

(2)
can be a significant source of SH$^+$ (e.g., Abel et al. 2008).

The strongest ground state FS component of SH$^+$($^3\Sigma^-$) at $\sim$526 GHz ($N_J = 1_0 - 0_1$) cannot be observed from the ground because of the water in Earth’s atmosphere. The $\sim$683 GHz ($N_J = 1_0 - 0_1$) and $\sim$893 GHz ($N_J = 2_1 - 1_1$) transitions can be observed from the ground, albeit with some difficulty. Transitions of the $N_J = 0_0 - 0_1$ FS component at $\sim$346 GHz (Savage et al. 2004) lie in a more accessible frequency window. Indeed, Menten et al. (2011) searched for the SH$^+$($^1\Sigma^-$) ($N_J = 1_0 - 0_1$, $F = 1/2 - 3/2$) transition in the Orion Bar between 345.8 GHz and 345.9 GHz. Blue arrows show the predicted frequencies of the SH$^+$($^1\Sigma^-$) transitions can be computed from the new experimental measurements Nyquist-sampled fields and provided a typical resolution of 3.6$''$. The total observing time was about 6.5 h and 2.2 h for ACA and ALMA observations respectively. The data were calibrated within the CASA software package using the standard

$$2.1. \text{Single-dish IRAM 30 m observations}$$

IRAM 30 m telescope observations were taken in 2013 using the EMIR330 receiver and the FFTS backend in the wide mode that covers a 16 GHz instantaneous bandwidth per polarization at a channel spacing of 195 kHz. They are part of a complete double-dish IRAM 30 m observations. They belong to the 2012.1.00352.S project (ACA) is used to complement ALMA observations of extended sources by producing short-spacing visibilities filtered out by the ALMA array. At the time of observations, the ACA array used nine 7 m antennas. The ACA mosaic contained ten pointings Nyquist-sampled fields and provided a typical resolution of 3.6$''$. The total observing time was about 6.5 h and 2.2 h for ACA and ALMA observations respectively. The data were calibrated within the CASA software package using the standard

$$2.2. \text{Interferometric ALMA and ACA observations}$$

Atacama Large Millimeter Array (ALMA) observations of the Orion Bar were carried out in 2013 and 2014 during ALMA Cycle 1 observations. They belong to the 2012.1.00352.S project “The fundamental structure of molecular cloud edges: from clumps to photoevaporation” (P.I.: I.R. Goicoechea). They consist of mosaicking observations covering a $\sim$40”x40” field centered at $\alpha_{2000} = 05^h35^m20.6^s$, $\delta_{2000} = -05^\circ25'20.0''$. The ALMA array observations used twenty-seven 12 m antennas. The field of view was observed as a mosaic of 27 Nyquist-sampled pointings in the C32-2 configuration, providing a typical resolution of $\sim$1”. Observations were carried out with the band 7 receiver near 346 GHz using the ALMA correlator backend at a spectral resolution of 488.3 kHz ($\sim$0.4 km s$^{-1}$) over a total bandwidth of 937.5 MHz. Atacama Compact Array (ACA) was used to complement ALMA observations of extended sources by producing short-spacing visibilities filtered out by the ALMA array. At the time of observations, the ACA array used nine 7 m antennas. The ACA mosaic contained ten pointings Nyquist-sampled fields and provided a typical resolution of $\sim$3.6”. The total observing time was about 6.5 h and 2.2 h for ACA and ALMA observations respectively. The data were calibrated within the CASA software package using the standard
The calibrated uv tables were then exported to GILDAS, where the data were deconvolved using the Högbom CLEAN algorithm. All technical details on the data reduction and the complete data set will be presented elsewhere. For the purpose of this work (showing the 345.8–345.9 GHz window observed by different instrumentation) we only deconvolved the ACA and ALMA visibilities separately.

3. Observational results

Figure 1 shows, from top to bottom, the IRAM 30 m, ACA, and ALMA spectra toward the Orion Bar dissociation front; we note that ACA and ALMA spectra are averaged over the same region of the dissociation front. The abscissa axis represents the rest wavelength of the dissociation front. It agrees, within ~0.2 km s⁻¹, with typical velocity centroids of other reactive molecular ions observed with single-dish telescopes toward the Orion Bar (see, e.g., Fuente et al. 2003; Leurini et al. 2006; Nagy et al. 2013).

We expected to detect the SH⁺ transition: \( F' = 1/2 \rightarrow 1/2 \) and \( F' = 1/2 \rightarrow 3/2 \) HFS lines of the \( N' = 1 \rightarrow 0 \) FS component near 346 GHz on the basis of the previous Herschel/HIFI detection of the higher energy SH⁺ \( N' = 1 \rightarrow 1 \) lines at ~526 GHz toward the Orion Bar (Nagy et al. 2013). Blue arrows in Fig. 1 show the absence of emission at frequencies derived from previous laboratory data of Savage et al. (2004) for all three telescopes. Our spectra, however, show two emission lines shifted by ~15 MHz (~13 km s⁻¹) to higher frequencies with respect to the frequencies indicated by blue arrows, which could not be assigned to other carriers in the MADEX, CDMS (Müller et al. 2001, 2005), and JPL (Pickett et al. 1998) spectroscopic catalogs. Table 1 shows results from Gaussian fits to the two lines, which were detected with up to 12\( \sigma \) and 7\( \sigma \) significance levels. Averaging the three observations, we obtain 345944.35 MHz and 345858.27 MHz for the two lines. Given the complex gas kinematics in Orion, implying an uncertainty in \( v_{\text{LSR}} \), we adopted a 1\( \sigma \) systematic uncertainty of 0.2 MHz for the rest frequency determination. We note that the line near 345.9 GHz is only ~61 MHz (~53 km s⁻¹) above the CO \( J = 3 \rightarrow 2 \) line frequency.

4. New SH⁺ spectroscopic parameters

The two unpaired electrons (\( S = 1 \)) of sulfanylium lead to a \( ^3\Sigma^− \) electronic ground state which causes all rotational levels \( N \) to be split into three with \( J = N, N \pm 1 \), except for the \( N = 0 \) level for which only \( J = 1 \) exists. The lowest order FS parameters are \( \lambda \) and \( \gamma \) which describe the (electron) spin-spin coupling and the (electron) spin-rotation coupling. Rotational (or centrifugal distortion) and vibrational corrections, the latter for excited vibrational states, may appear in the Hamiltonian. The proton magnetic moment \( (I_H = 1/2) \) splits every FS level into two HFS levels. The main HFS parameters here are \( b_y \) and \( c \), the scalar and tensorial electron spin-nuclear spin coupling parameters.

The selection rules \( \Delta F = 0, \pm 1 \) hold strictly. The spin-conserving transitions (\( \Delta F = \Delta J = \Delta N \)) are the strongest ones at higher quantum numbers. Other transitions have non-negligible intensities, in particular at low-\( N \). The \( N = 1 \rightarrow 0 \) ground state rotational transition is split into three FS components \( J = 0 \rightarrow 1, 2 \rightarrow 1, \) and \( 1 \rightarrow 1 \) near 346, 526, and 683 GHz, respectively, which are split further into 2, 3, and 4 HFS components. An energy level diagram is shown, e.g., in Menten et al. (2011).

The rotation-vibration spectrum of SH⁺ was recorded up to \( \nu = 2 \rightarrow 1 \) (Brown et al. 1986) and \( \nu = 4 \rightarrow 3 \) (Civiš et al. 1989). Hovde & Saykally (1987) investigated the rotational spectrum of SH⁺ in its ground and first excited vibrational state between 0.51 THz and 1.62 THz by laser magnetic resonance (LMR). More recently, Savage et al. (2004) employed source-and-velocity-modulation to determine accurate rest frequencies of all three HFS components near 526 GHz of the \( N = 1 \rightarrow 0 \) transition of SH⁺ as well as one of two HFS components near 346 GHz. In each of these four studies, spectroscopic parameters were determined only from their own data. In contrast, Brown & Müller (2009) used the field-dependent (Hovde & Saykally 1987) and field-free (Savage et al. 2004) rotational data as well as the rovibra- tional data of the fundamental (\( \nu = 1 \rightarrow 0 \)) band (Brown et al. 1986; Civiš et al. 1989) to determine ground state spectroscopic parameters as well as vibrational corrections if they were needed. One of the present authors (HSPM) used a slightly different approach to create an entry for the CDMS catalog because the SPFIT program (Pickett 1991) does not permit the use of field-dependent data. The LMR transition frequencies extrapolated to zero field by Brown & Müller (2009) were fitted together with the field-free rotational data (Savage et al. 2004) and all of the rovibra- tional data (Brown et al. 1986; Civiš et al. 1989) to determine Dunham-type parameters \( P_{ij} \) for SH⁺, with \( i \) and \( j \) indicating vibrational and rotational corrections, respectively.

| Parameter | Value |
|-----------|-------|
| \( Y_{10} \) | 2.5474948 (104) |
| \( Y_{20} \) | −49.4293 (90) |
| \( Y_{00} \) | 0.2097 (30) |
| \( Y_{20} \times 10^{3} \) | −16.01 (34) |
| \( Y_{01} \) | 278.0949 (36) |
| \( Y_{11} \) | −8.5773 (85) |
| \( Y_{21} \) | 16.15 (32) |
| \( Y_{02} \) | −14.7380 (76) |
| \( Y_{12} \times 10^{3} \) | 122.9 (27) |
| \( Y_{03} \times 10^{3} \) | 0.46 |
| \( A_{00} \) | 171.4883 (58) |
| \( A_{10} \) | −471.8 (146) |
| \( A_{20} \) | −79.1 (67) |
| \( A_{01} \) | −1.13 (24) |
| \( A_{00} \) | −5.0369 (91) |
| \( A_{10} \) | 116.4 (20) |
| \( A_{20} \) | 3.52 (64) |
| \( A_{01} \) | 0.432 (35) |
| \( b_{F0} (1H) \) | −56.884 (81) |
| \( b_{F1} (1H) - b_{F0} (1H) \) | −3.46 (79) |
| \( c (1H) \) | 33.60 (67) |

\( a \) Numbers in parentheses are one standard deviation in units of the least significant digits.

\( b \) In units of \( \text{cm}^{-1} \).
Savage et al. (2004) observed all three HFS components of the $N_J = 1_2 - 0_1$ transition of sulfanylium (around 526.1 GHz) between 0.2 MHz and 0.5 MHz higher than calculated from the parameters of the LMR study (Hovde & Saykally 1987); the deviations are well within the uncertainties of $\sim$1.6 MHz from that study. These rest frequencies improve the accuracies of the major HFS parameters $b_F$ and $c$, as well as the origin of the FS component, which depends on several rotational and fine structure parameters. The $F = 1/2 - 3/2$ HFS component was found by Savage et al. (2004) only 1.23 MHz higher than calculated from parameters in Hovde & Saykally (1987), well within the uncertainty of $\sim$18 MHz. The uncertainty is much larger because this FS component was not accessed in the LMR study. The associated $F = 1/2 - 1/2$ HFS component, weaker by a factor of two, was not detected despite significant signal averaging. Its frequency, $\sim$86 MHz lower than the $F = 1/2 - 3/2$ HFS component, is well constrained by the frequency of this HFS component and from the data near 526.1 GHz.

As outlined in section 3, no emission lines were detected in the Orion Bar at the frequencies expected from Savage et al. (2004). However, the two emission lines, each observed $\sim$15 MHz higher, had the expected 1:2 intensity ratio and spacing required to assign them to the $N_J = 1_2 - 0_1$ FS component. In order to test the feasibility of these assignments, we omitted the laboratory transition frequency near 346 GHz from the data set which was used to create the CDMS catalog entry. Interestingly, the rms error of the fit improved significantly from 0.898 to 0.826. More importantly, the two HFS components were now predicted at 345856.8 MHz and at 345943.0 MHz, very close to our observed emission lines (Fig. 1) and each with predicted 1σ uncertainties of 3.9 MHz. These uncertainties are considerably smaller than those from Hovde & Saykally (1987), mainly because of the extensive rotational vibrational transition frequencies used in the present fit and because of the remaining data from Savage et al. (2004). Hence, we used averages over three independent observations given in section 3 to redetermine the SH$^+$ spectroscopic parameters. The resulting parameters are given in Table A1. The rms error of this fit is 0.820, demonstrating that our transition frequencies are more compatible with the remainder of the laboratory data than the single frequency near 346 GHz determined by Savage et al. (2004). Updated predictions of the rotational spectrum of SH$^+$ will be available in the CDMS catalog. Data up to 1 THz are given in Table A2.

Errors in rest frequencies determined in the laboratory or through astronomical observations are known to occur especially in sparse data sets. Recent error correction of data pertaining to astrophysically important hydrides include the correction of the fundamental transition of HD$^+$ (Asvany et al. 2008) and of the fundamental transitions of CH$^+$ isotopologues (Amano 2010).

The changes in spectroscopic parameters are small for the most part and occur predominantly in rotational and FS parameters. The most significant changes of 3σ occur in $V_01 \approx V_02$, and $V_03 \approx -D_0$. The largest change in magnitude occurs in $\lambda_{02}$, but corresponds only to $\sim$1σ. The strong, spin-conserving transitions are only slightly affected at lower frequencies. In addition, the transitions of the $N_J = 1_1 - 0_0$ FS component (3683 GHz) are now predicted 2 MHz lower than in the first SH$^+$ CDMS catalog entry. Changes from the predictions by Brown & Müller (2009) are a bit more complex, but deviate on average by $\sim$2 MHz. Larger deviations of several megahertz may occur for transitions with flip of the electron spin (e.g., at $\sim$893 GHz) or at higher frequencies.

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Brünnken et al. (2014), in particular in combination with a study of 34SH$^+$.

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Table A.1. Gaussian fits to the $F = 0.5 - 1.5$ and $0.5 - 0.5$ lines of the $N_J = 1_0 - 0_1$ FS component of SH$^+$ observed with three different telescopes toward the Orion Bar (for $v_{LSR}=10.50\,\text{km}\,\text{s}^{-1}$).

| Telescope   | Line       | Rest frequency (MHz) | $\int T_{mb}dv$ (mK km s$^{-1}$) | Line width (km s$^{-1}$) | Signal-to-noise |
|-------------|------------|----------------------|----------------------------------|--------------------------|-----------------|
| IRAM 30 m   | 1.5 − 0.5  | 345944.22 (20)       | 383 (30)                         | 1.95 (19)                | ~10             |
|             | 0.5 − 0.5  | 345858.38 (20)       | 185 (31)                         | 1.68 (32)                | ~5              |
| ACA         | 1.5 − 0.5  | 345944.55 (20)       | 260 (20)                         | 1.23 (10)                | ~12             |
|             | 0.5 − 0.5  | 345858.42 (20)       | 114 (25)                         | 1.10 (26)                | ~6              |
| ALMA        | 1.5 − 0.5  | 345944.29 (20)       | 668 (51)                         | 1.20 (10)                | ~12             |
|             | 0.5 − 0.5  | 345858.00 (20)       | 295 (58)                         | 0.92 (20)                | ~7              |

**Notes.** Parentheses show the Gaussian fit uncertainties (in units of the least significant digits). For the fitted frequencies, we adopt an overall 1σ uncertainty of 0.2 MHz (~0.2 km s$^{-1}$). This should be considered as a systematic uncertainty due to uncertainties in $v_{LSR}$ (see text). IRAM 30 m spectrum smoothed to ~0.4 km s$^{-1}$, similar to ALMA and ACA; Signal-to-noise determined on the peaks and referring to ~0.4 km s$^{-1}$ resolution for IRAM 30 m and to the native spectral resolution for ALMA and ACA.

Table A.2. Quantum numbers, frequencies (MHz), uncertainties unc. (MHz), Einstein $A$ values ($10^{-4}$ s$^{-1}$), upper $g_{up}$ and lower $g_{lo}$ state degeneracies, and upper $E_{up}$ and lower $E_{lo}$ state energies (cm$^{-1}$) of sulfanylium, SH$^+$, below 1 THz.

| $N' - N''$ | $J' - J''$ | $F' - F''$ | Frequency | unc. | $A$ | $g_{up}$ | $g_{lo}$ | $E_{up}$ | $E_{lo}$ |
|------------|-------------|-------------|-----------|------|-----|----------|----------|----------|----------|
| 1 − 0      | 0 − 1       | 0.5 − 0.5   | 345858.1957 | 0.1494 | 1.15 | 2        | 2        | 11.5395  | 0.0029   |
| 1 − 0      | 0 − 1       | 0.5 − 1.5   | 345944.4205 | 0.1496 | 2.30 | 2        | 4        | 11.5395  | 0.0000   |
| 1 − 0      | 2 − 1       | 1.5 − 0.5   | 526038.7348 | 0.0699 | 8.00 | 4        | 2        | 17.5496  | 0.0029   |
| 1 − 0      | 2 − 1       | 2.5 − 1.5   | 526047.9440 | 0.0714 | 9.59 | 6        | 4        | 17.5471  | 0.0000   |
| 1 − 0      | 2 − 1       | 1.5 − 1.5   | 526124.9597 | 0.0723 | 1.60 | 4        | 4        | 17.5496  | 0.0000   |
| 1 − 0      | 1 − 1       | 1.5 − 0.5   | 683334.0618 | 0.5139 | 2.90 | 2        | 4        | 22.7964  | 0.0029   |
| 1 − 0      | 1 − 1       | 0.5 − 0.5   | 683359.9250 | 0.4790 | 11.59 | 2        | 2        | 22.7973  | 0.0029   |
| 1 − 0      | 1 − 1       | 1.5 − 1.5   | 683420.2867 | 0.4972 | 14.48 | 4        | 4        | 22.7964  | 0.0000   |
| 1 − 0      | 1 − 1       | 0.5 − 1.5   | 683446.1499 | 0.4942 | 5.79  | 2        | 4        | 22.7973  | 0.0000   |
| 2 − 1      | 1 − 1       | 0.5 − 0.5   | 893065.8800 | 0.9514 | 19.77 | 2        | 2        | 52.5868  | 22.7973  |
| 2 − 1      | 1 − 1       | 0.5 − 1.5   | 893091.7433 | 0.9763 | 9.89  | 2        | 4        | 52.5868  | 22.7964  |
| 2 − 1      | 1 − 1       | 1.5 − 0.5   | 893126.2353 | 1.0505 | 4.94  | 4        | 2        | 52.5888  | 22.7973  |
| 2 − 1      | 1 − 1       | 1.5 − 1.5   | 893152.0985 | 0.9475 | 24.73 | 4        | 4        | 52.5888  | 22.7964  |