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Fe^+ IN ITS GROUND ^6D STATE: A DIRECT MEASUREMENT OF THE $J = 1/2$–$3/2$ AND $J = 3/2$–$5/2$ FINE-STRUCTURE INTERVALS[1]

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ABSTRACT

The $J = 1/2$–$3/2$ and $3/2$–$5/2$ fine-structure transitions in the atomic ion Fe^+ in its ground ^6D state have been detected in the laboratory by far-infrared laser magnetic resonance at 87 and 51 μm. The fine-structure intervals have been measured accurately by extrapolation to zero magnetic field as 3430.7762 (19) and 5843.7984 (12) GHz, respectively.

Subject headings: atomic data — infrared: general — line: identification — methods: laboratory

1. INTRODUCTION

The lowest electronic configuration of Fe^+ is $3d^64s^1$, which gives rise to an inverted ^6D ground state; the levels of this state are shown in Figure 1. The fine-structure transitions fall in the far-infrared region of the spectrum, with wavelengths for the successive intervals of 25.99, 35.35, 51.30, and 87.38 μm (the first of these intervals is $J = 7/2$–$9/2$ and so on). So far these intervals have only been measured indirectly from optical spectra, with an uncertainty of a few hundred megahertz (Corliss & Sugar 1982). In this paper, we report the direct laboratory observation of the $J = 3/2$–$5/2$ and $1/2$–$3/2$ fine-structure transitions which are magnetic dipole in character. The observations were made by laser magnetic resonance (LMR) in the far-infrared and yield accurate transition frequencies of 5843798.4 ± 1.2 MHz and 3430776.2 ± 1.9 MHz.

Atomic fine-structure transitions are useful probes of the local physical conditions in various astrophysical situations. For example, the fine-structure transitions in C^+ (^2P) at 158 μm and in N^+ (^3P) at 122 and 205 μm are prominent in emission from the gaseous component of the interstellar medium (Wright et al. 1991); the signals provide information on the large-scale structure of our Galaxy. Similarly, the fine-structure transition in S (^3P) at 25.2 μm provides an important diagnostic for interstellar shocks (Tielens & Hollenbach 1985; Haas, Hollenbach, & Erickson 1991). The fine-structure intervals of light atoms fall in the far-infrared region of the electromagnetic spectrum. Many of them have been determined with an accuracy of ~ 1 GHz from differences between lines in optical spectra (Moore 1949). In recent years, such transitions have been detected directly in astrophysical sources by either heterodyne or Fabry-Perot interferometric techniques. The observations reveal a need for more accurate determinations of the transition frequencies. If the observation is to be used to measure, for example, the relative velocities of different components of interstellar gas, the rest frequency needs to be known to within a few megahertzs as shown by the measurements on C^+ by Lutgen et al. (1986). A program to measure these intervals by far-infrared LMR spectroscopy has been established at the Boulder laboratories of the National Institute of Standards and Technology.

The element iron has a relatively high cosmic abundance of 0.003% (Rowan-Robinson 1981). For this reason, not only should it be detectable in astronomical sources, but it is also likely to play a significant part in astrophysical processes. Several observations of the ion Fe^+ have already been made in remote sources. For example, lines of Fe^+ in the ultraviolet have been detected in the diffuse interstellar medium (ISM), looking toward ζ Oph by Morton (1975). Analysis of these observations showed that Fe is about 2 orders of magnitude less abundant in the ISM than in the Sun. More recently, forbidden transitions of Fe^+ (between quartet and sextet states) have been detected in emission in the near-infrared by Simpson et al. (1996). These observations suggest that the dominant mechanism of excitation of the line-emitting gas in "active" galaxies is through photoionization of material with normal ISM abundance.

2. EXPERIMENTAL DETAILS

The LMR spectrometer used in this work has been described elsewhere (Sears et al. 1982), and the details are not repeated here. As described in earlier papers on atomic fine-structure transitions (Brown, Zink, & Evenson 1994a), we have recently increased its sensitivity by raising the modulation frequency from 13 to 40 kHz. In addition, we have modified the spectrometer to enhance its performance at wavelengths shorter than 100 μm by reducing the inside diameter of the polished copper laser-pump tube from 50.8 to 19.1 mm (from 2 to $\frac{3}{4}$ inches). This provides much better overlap between the pumped lasing gas and the far-infrared radiation field within the laser cavity, permitting many more short-wavelength laser lines to oscillate. In particular, we have used one such line of CD_3OH to study the $J = 3/2$–$5/2$ fine-structure transition in Fe^+. This line at 51.5 μm has not been reported previously, but it lases readily in the new arrangement. In order to use it in the study of Fe^+, we have determined its frequency by measuring the beat frequency when mixed with a pair of CO_2 laser frequencies in a metal-insulator-metal (MIM) diode. The result is as follows: CD_3OH pump $10R$ (56), $\lambda = 51.48$ μm, $\nu = 5823660.9 \pm 0.7$ MHz. This frequency lies about 20 GHz below that of the $J = 3/2$–$5/2$ transition in Fe^+ and is

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well within the tuning range of our magnet (maximum flux density of 2.1 T). The $J = 1/2$–$3/2$ transition occurs at lower frequencies but also requires short-wavelength laser lines, below 100 μm. The transition has been detected with two laser lines at 86.74 and 86.24 μm, both of which have been frequency measured previously (Douglas 1989).

The Fe^+ atoms were generated in the gas phase by using the same microwave discharge source as we used to form N^+ ions (see Brown et al. 1994b). The discharge was run through ultra–high-purity helium at 133 Pa (1 Torr) with a small amount of iron pentacarbonyl vapor, $Fe(CO)_5$, added; the optimum pressure of the $Fe(CO)_5$ was 0.13 Pa (1m Torr).

3. RESULTS, ANALYSIS, AND DISCUSSION

The LMR spectra associated with the $J = 1/2$–$3/2$ and $J = 3/2$–$5/2$ fine-structure transitions of Fe^+ in its ground ^6D state have been recorded. The energy-level scheme is shown in Figure 1. The Landé g-factors decrease with increasing J-value. It is therefore possible to detect transitions in both parallel ($\boldsymbol{B}_{laser} \parallel \boldsymbol{B}_{lab}$) and perpendicular ($\boldsymbol{B}_{laser} \perp \boldsymbol{B}_{lab}$) polarizations. However, the parallel transitions, with $\Delta M_J = 0$, tune more slowly, too slowly in fact for us to be able to detect them with the laser lines used in these experiments. Examples of some of the spectra recorded in absorption are shown in Figure 2 ($J = 1/2$–$3/2$) and Figure 3 ($J = 3/2$–$5/2$. It is worth noting that the lower levels of these transitions are 863 and 668 cm^{-1}, respectively, above the ground $J = 9/2$ level (Corliss & Sugar 1982). The observed signals arise predominantly from $^{56}Fe^+$, which has a natural abundance of 91.7%. The other two significant isotopes of Fe probably contribute to the signal also because neither the isotope shift (in the case of

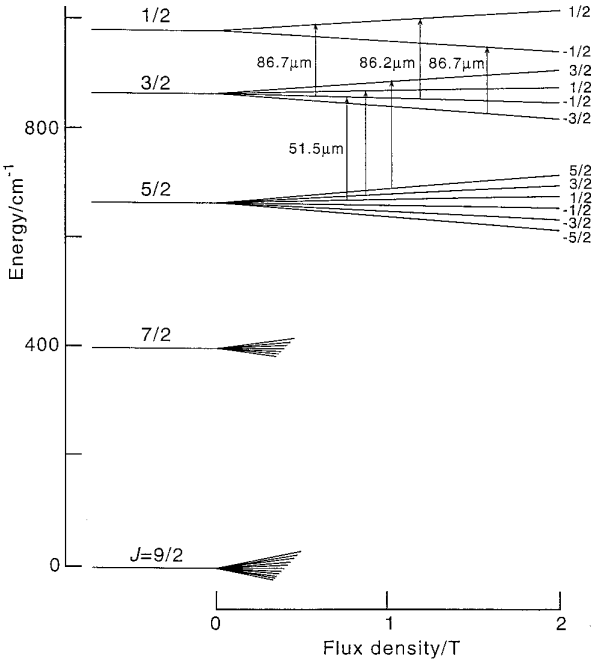

FIG. 1.—Energy-level diagram for the ground ^6D state of the atomic ion Fe^+ in the presence of a variable magnetic field. The Zeeman components are labeled by their M_J values; the Zeeman splittings are exaggerated for the sake of clarity. The three transitions involving the $J = 1/2$, 3/2, and 5/2 levels, which were observed in the present work, are indicated with their laser wavelength.

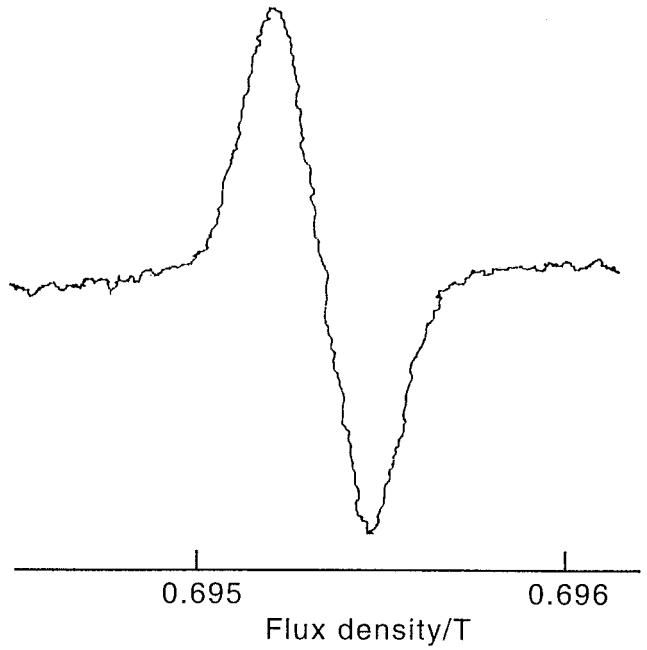

FIG. 2.—Part of the far-infrared LMR spectrum associated with the $J = 1/2$–$3/2$ transition of Fe^+ in its ground ^6D state, recorded with the 86.74 μm line of CD_3OH, pumped by the 10R (36) line of a CO_2 laser. The spectrum was recorded with the oscillating magnetic field perpendicular to the DC magnetic field ($\Delta M_J = \pm 1$). The transition involved is $M_J = 1/2$ to $-1/2$. The output time constant for the lock-in amplifier was 0.3 s.

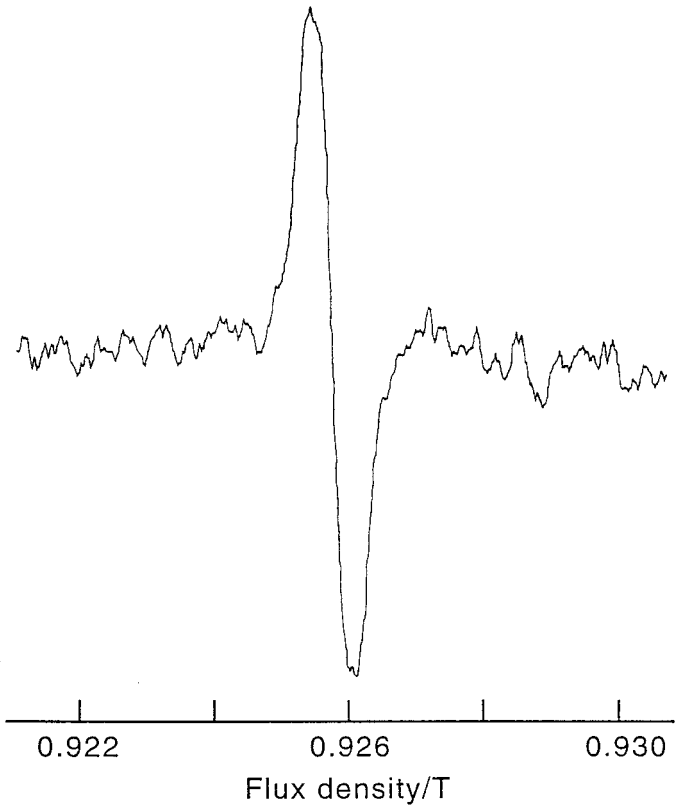

FIG. 3.—Part of the far-infrared LMR spectrum associated with the $J = 3/2$–$5/2$ transition of Fe^+ in its ground ^6D state, recorded with the 51.48 μm line of CD_3OH, pumped by the 10R (56) line of a CO_2 laser. The spectrum was recorded in perpendicular polarization for magnetic dipole transitions ($\Delta M_J = \pm 1$). The transition involved is $M_J = 1/2$–$3/2$, and the output time constant for the lock-in amplifier was 0.3 s.

TABLE 1

LASER MAGNETIC RESONANCE DATA FOR Fe^+ IN ITS GROUND ^6D STATE

J	M_J	ν_L (GHz	B_0 (mT)	$o-c$ (MHz)
$\frac{1}{2} \leftarrow \frac{3}{2}$.....	$\frac{1}{2} \leftarrow -\frac{1}{2}$	3456.1612[a]	695.36	−0.2
	$-\frac{1}{2} \leftarrow -\frac{3}{2}$	3456.1612	1591.42	0.0
	$\frac{1}{2} \leftarrow -\frac{1}{2}$	3476.2825[b]	1244.09	0.1
$\frac{3}{2} \leftarrow \frac{5}{2}$.....	$-\frac{1}{2} \leftarrow \frac{1}{2}$	5823.6609[c]	815.78	−0.3
	$\frac{1}{2} \leftarrow \frac{3}{2}$	5823.6609	925.43	0.5
	$\frac{3}{2} \leftarrow \frac{5}{2}$	5823.6609	1072.50	−0.2

[a] The 86.74 μm line of CD_3OH, pumped by the 10R(34) line of CO_2.
[b] The 86.24 μm line of CH_3OH, pumped by the 9R(08) line of CO_2.
[c] The 51.48 μm line of CD_3OH, pumped by the 10R(56) line of CO_2.

^{54}Fe, 5.8% abundance) nor the magnetic hyperfine splitting (in the case of ^{57}Fe, 2.2% abundance) is large enough to shift the signal for these isotopes outside the main line shape. The magnetic moment of the ^{57}Fe nucleus is very small, $0.091\mu_N$.

The LMR spectra of Fe^+ have been analyzed with a standard effective Hamiltonian for a Russell-Saunders atom, as described for example by Cooksy et al. (1986). The observations depend directly on five parameters, the fine-structure intervals for $J = 1/2$–$3/2$ and $3/2$–$5/2$ and the three g-factors for the levels $J = 1/2$, $3/2$, and $5/2$. Values for these five parameters have been determined in the fit of the six measurements listed in Table 1. The results are given in Table 2, together with the other parameters in the model which were constrained in the fit. The values for the five determined parameters are consistent with those determined previously from optical measurements but are more accurate; see Table 2. It turns out that for Fe^+ (and Fe) atoms, some extremely accurate measurements of the ultraviolet spectrum have been made recently by Fourier transform spectrometry (Nave et al. 1991). The purpose of these measurements was to provide improved calibration standards in this region of the spectrum; the relative precision of the measurements is 3×10^{-8}, and the estimated absolute accuracy is 0.002 cm^{-1} or 60 MHz. We have used these measurements to determine the fine-structure intervals for Fe^+. The values, which are also given in Table 2, are in remarkably good agreement with our direct measurements. The g-factors of Fe^+ were known only approximately before so, even though there are limitations to the accuracy with which we can measure them by FIR LMR spectroscopy (Brown, Zink, & Evenson 1998), we have been able to improve on them. It is interesting to note that the values

TABLE 2

PARAMETERS DETERMINED FROM THE FAR-INFRARED LMR SPECTRUM OF $^{56}Fe^+$ ATOMS

Parameter	This Work	Previous Values
$(E_{1/2} - E_{3/2})$ GHz^{-1}	3430.7762(19)[a]	3430.82[b], 3430.8114[c]
$(E_{3/2} - E_{7/2})$ GHz^{-1}	5843.7984(12)	5843.85[b], 5843.7689[c]
$(E_{5/2} - E_{7/2})$ GHz^{-1}		8480.92[b,d], 8481.0135[c]
$(E_{7/2} - E_{9/2})$ GHz^{-1}		11535.71[b,d], 11535.6203[c]
$g_{J=1/2}$	3.33623(31)	3.31[b], 3.333[e]
$g_{J=3/2}$	1.86753(10)	1.862[b], 1.867[e]
$g_{J=5/2}$	1.65772(20)	1.655[b], 1.657[e]
$g_{J=7/2}$		1.58[b,d], 1.587[e]
$g_{J=9/2}$		1.58[b,d], 1.556[e]

[a] Number in parentheses represents 1 σ uncertainty estimates, in units of the last quoted decimal place.
[b] Values determined from optical spectra given by Corliss & Sugar 1982.
[c] Value calculated from the Fourier-transform recording of the ultraviolet spectrum of Fe^+ by Nave et al. 1991.
[d] Parameter constrained to this value in the least-squares fit.
[e] Value calculated from the Landé g-factor formula.

estimated from the Landé g-factor formula agree better with the present measurements than do the old experimental values (see also Table 2).

We hope that the accurate measurements of the $J = 1/2$–$3/2$ and $J = 3/2$–$5/2$ fine-structure intervals in Fe^+ at 87.4 and 51.3 μm will be of use for the detection of this atomic ion in a wider range of astrophysical sources. However, it is desirable to measure the higher frequency fine-structure intervals as well because they will probably be more useful for astronomical purposes. These transitions involve the lower spin components with a greater population factor (see Fig. 1), and they have larger Einstein A-coefficients because of the ν^3 factor. The Einstein A-coefficients for the four fine-structure transitions are given in Table 3. In principle, the higher frequency intervals can also be measured in an LMR experiment. However, they lie outside the range of our photoconductive detector as configured at present, and appropriate far-infrared laser lines would need to be discovered before the experiment could be attempted. The Einstein B-coefficient is a better measure of laboratory line strength; at far-infrared wavelengths, the absorption intensity is proportional to $\nu^2 B_{ij}$. The values for B_{ij} and $\nu^2 B_{ij}$ for the four fine-structure transitions of Fe^+ are also given in Table 3.

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TABLE 3

EINSTEIN A- AND B-COEFFICIENTS FOR FINE-STRUCTURE TRANSITIONS IN ATOMIC Fe^+ IN ITS GROUND ^6D STATE[a]

Transition	Frequency (GHz)	$10^4 A_{ij}$(s^{-1})	$10^{-16} B_{ij}$(kg^{-1} m)	$10^{-41} \nu^2 B_{ij}$(kg^{-1} ms^{-2})
$^6D_{1/2} - {}^6D_{3/2}$.....	3430.776	1.895	2.386	2.808
$^6D_{3/2} - {}^6D_{5/2}$.....	5843.798	7.225	2.454	8.380
$^6D_{5/2} - {}^6D_{7/2}$.....	8481.014	15.775	1.972	14.182
$^6D_{7/2} - {}^6D_{9/2}$.....	11535.620	21.442	1.136	15.117

[a] A- and B-coefficients have been calculated in the Russell-Saunders limit using the formula given by Corney 1979.

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