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ATOMIC IRON IN ITS ^5D GROUND STATE: A DIRECT MEASUREMENT OF THE $J = 0 \leftarrow 1$ AND $J = 1 \leftarrow 2$ FINE-STRUCTURE INTERVALS[1,2]

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ABSTRACT

The $J = 0 \leftarrow 1$ and $J = 1 \leftarrow 2$ fine-structure transitions in atomic Fe in its ground ^5D state have been detected in the laboratory by far-infrared laser magnetic resonance. The fine-structure intervals have been measured accurately as 2696.3942(22) and 5519.9397(18) GHz, respectively; these values correspond to wavelengths of 111.18273 and 54.31082 μm, with an uncertainty of about 2×10^{-5} μm.

Subject headings: atomic data — ISM: atoms — line: identification — radio lines: ISM

1. INTRODUCTION

It is estimated that iron has a relatively high cosmic abundance, 0.003% (Rowan-Robinson 1981). There is therefore a good chance that this element is detectable in astronomical sources, either as atoms or in combination with other elements. Indeed, several of the dark lines observed almost 200 years ago in the solar spectrum by Fraunhofer (1817) were assigned, after a respectable interval, to transitions in atomic iron by Kirchhoff (1859). In more recent times, the diatomic molecule FeH has been identified in the atmospheres of cool stars of spectral types M, S, and K, including our Sun (Carroll & McCormack 1972; Carroll, McCormack, & O'Connor 1976). Searches have also been made with a radio telescope for FeO in the interstellar medium, but so far the results have been negative (Merer, Walmsley, & Churchwell 1982).

Astronomers have made extensive use of heterodyne and Fabry-Perot receivers on airborne platforms in recent years. These measurements have been used both to identify interstellar species and to determine the relative velocities of different components of the interstellar gas. Such experiments require an accurate knowledge of their rest frequencies, valid to a few megahertz. Far-infrared laser magnetic resonance (LMR) spectroscopy has proved itself to be a powerful method for obtaining these frequencies in the laboratory.

The lowest electronic configuration of atomic Fe is $3d^64s^2$, which gives rise to an inverted ^5D ground state. The levels of this state are shown in Figure 1. The fine-structure separations fall in the far-infrared region of the spectrum, with the wavelengths for the successive intervals of 24.0, 34.7, 54.3, and 111.2 μm (the first of these intervals is $J = 3 \leftarrow 4$ 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 *Letter* we report the first laboratory detection of the weak, magnetic dipole $J = 0 \leftarrow 1$ and $J = 1 \leftarrow 2$ transitions in Fe in its ground state, which has allowed us to measure their frequencies very accurately.

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 our earlier paper on atomic Si (Brown, Zink, & Evenson 1994), we have recently increased its sensitivity by raising the Zeeman modulation frequency from 13 to 40 kHz. We have also modified the spectrometer to enhance its performance at wavelengths shorter than 100 μm by reducing the inside diameter of the polished copper 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 $J = 1 \leftarrow 2$ fine-structure transition in Fe. This line has been reported previously by Sigg, Bluyssen, & Wyder (1984) but could not be made to lase in the previous configuration of the LMR spectrometer with the larger bore pump tube. It lased readily in the new arrangement. Since only its wavelength had been measured previously, we determined its frequency by measuring the beat frequency when mixed with a pair of CO_2 laser frequencies in a metal-insulator-metal diode. The results are as follows: CD_3OH pump $9P(28)$, $\lambda = 54.11$ μm, $\nu = 5540115.9 \pm 0.7$ MHz.

This frequency lies about 20 GHz above that of the $J = 1 \leftarrow 2$ transition in Fe and is well within the Zeeman tuning range of our magnet (maximum flux density of 2.1 T). The $J = 0 \leftarrow 1$ transition occurs at lower frequencies and can be studied with well-established laser lines at 110.4 and 110.7 μm.

The Fe atoms were generated in the gas phase by the reaction between H atoms and iron pentacarbonyl, $Fe(CO)_5$. The H atoms were produced by causing an approximately 2% mixture of hydrogen in helium to flow through a 2450 MHz discharge. The total pressure was about 200 Pa, and the

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[2] Contribution of US Government.

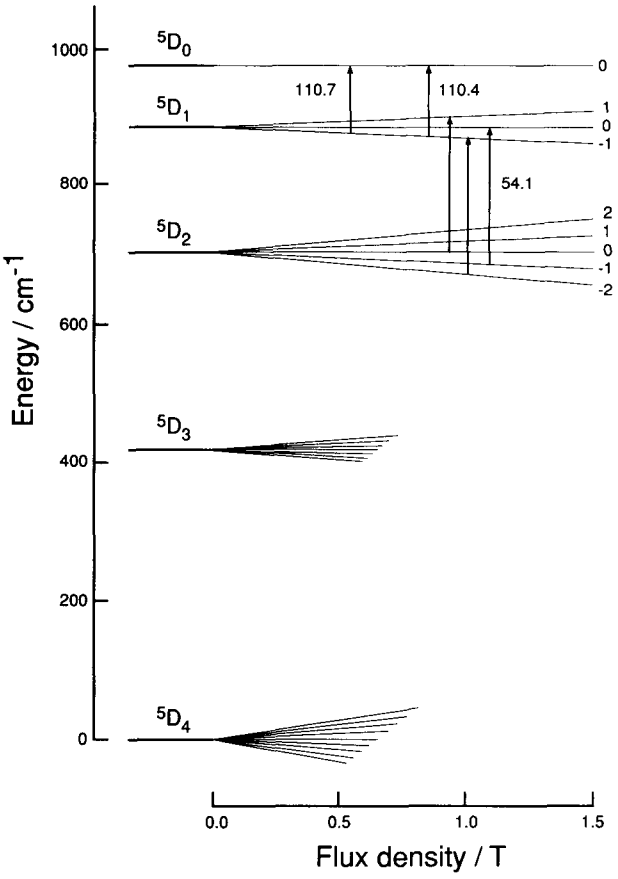

FIG. 1.—Energy-level diagram for the ground ^5D state of atomic iron, in the presence of a variable magnetic field. The Zeeman components are labeled by their M_J values: the Zeeman splittings are exaggerated for the sake of clarity. The three transitions involving the $J = 0$, 1, and 2 levels which were observed in the present work are indicated.

optimum Fe signal was obtained with a partial pressure of $Fe(CO)_5$ of about 0.3 Pa. This reaction had been used previously to generate FeH radicals in the gas phase (Beaton et al. 1988).

3. RESULTS, ANALYSIS, AND DISCUSSION

The LMR spectra associated with the $J = 0 \leftarrow 1$ and the $J = 1 \leftarrow 2$ fine-structure transitions of Fe in its ground ^5D state have been recorded. Since the Landé g-factors for all spin components of a ^5D state with $J \geq 1$ have the same value (1.5), the fine-structure transitions are significantly field-dependent only in perpendicular polarization (i.e., $B_{\rm laser} \perp B_{\rm lab}$) when they obey the selection rule $\Delta M_J = \pm 1$. The LMR spectrum for the $J = 0 \leftarrow 1$ transition consists of a single transition ($M_J = 0 \leftarrow -1$), whereas that for $J = 1 \leftarrow 2$ consists of three transitions, $M_J = -1 \leftarrow -2$, $0 \leftarrow -1$, and $1 \leftarrow 0$ with relative intensities 6:3:1; see Figure 1. Examples of the two spectra are shown in Figures 2 and 3. It is worth noting that the lower levels of these two transitions are 978 and 888 cm^{-1}, respectively, above the ground $J = 4$ 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 ^{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 signals from these isotopes outside the main line shape. The magnetic moment of the ^{57}Fe nucleus is very small, 0.091 nuclear magnetons.

The LMR spectra have been analyzed with a standard effective Hamiltonian for a Russell-Saunders atom, as described for example by Cooksy et al. (1986). The g-factors for atomic Fe in the ^5D state have been measured very accurately by Childs & Goodman (1965) in an atomic beam experiment. Because we cannot hope to match this accuracy in a far-infrared LMR experiment, we have constrained the g-factors to their atomic beam values in our fit. Childs & Goodman determined the g_J values for the individual J-levels for ^{57}Fe; in the case of ^{56}Fe, they were only able to determine a single value which was a population average over all the spin components. However, since the isotopic dependence of the g_J factors is much too small to be of significance for our measurements, we have assumed that the values quoted by Childs & Goodman (1965) for ^{57}Fe apply to all isotopes. Our data can then be used to determine much improved values for the $J = 0$–1 and $J = 1$–2 fine-structure intervals for atomic Fe; the results of the least-squares fit are given in Tables 1 and 2. The best values for the fine-structure intervals available previously were obtained indirectly from optical data (Corliss & Sugar 1982). These

TABLE 1

LASER MAGNETIC RESONANCE DATA FOR Fe IN ITS GROUND ^5D STATE

J	M_J	ν_L (GHz)	B_0 (mT)	$O-C$ (MHz)
$0 \leftarrow 1$.....	$0 \leftarrow -1$	2707.7493[a]	539.34	−1.2
	$0 \leftarrow -1$	2714.7147[b]	868.56	1.2
$1 \leftarrow 2$.....	$1 \leftarrow 0$	5540.1159[c]	957.98	2.7
	$-1 \leftarrow -2$	...	959.73	−2.9
	$0 \leftarrow -1$	...	964.28	0.3

[a] The 110.7 μm line of CH_3OH, pumped by the 9P(36) line of CO_2.
[b] The 110.4 μm line of $^{13}CH_3OH$, pumped by the 10R(18) line of CO_2.
[c] The 54.1 μm line of CD_3OH, pumped by the 9P(28) line of CO_2.

TABLE 2

PARAMETERS DETERMINED FROM FAR-INFRARED LMR SPECTRUM OF Fe ATOMS

Parameter	This Work	Previous Values
ΔE_{01}/GHz.....	2696.3942 (22)[a]	2696.42[c]
ΔE_{12}/GHz.....	5519.9397 (18)	5519.93[c]
ΔE_{23}/GHz.....	...	8636.18[b,c]
ΔE_{34}/GHz.....	...	12469.33[b,c]
$g_{J=1}$	...	1.50022[b,d]
$g_{J=2}$	...	1.50041[b,d]
$g_{J=3}$	...	1.50034[b,d]
$g_{J=4}$	...	1.50020[b,d]

[a] The numbers in parentheses represent 1 σ uncertainty estimates, in units of the last quoted decimal place.
[b] Parameter constrained to this value in the least-squares fit.
[c] Values determined from optical spectra, given by Corliss & Sugar 1982.
[d] Values determined for ^{57}Fe by atomic beam magnetic resonance (Childs & Goodman 1965).

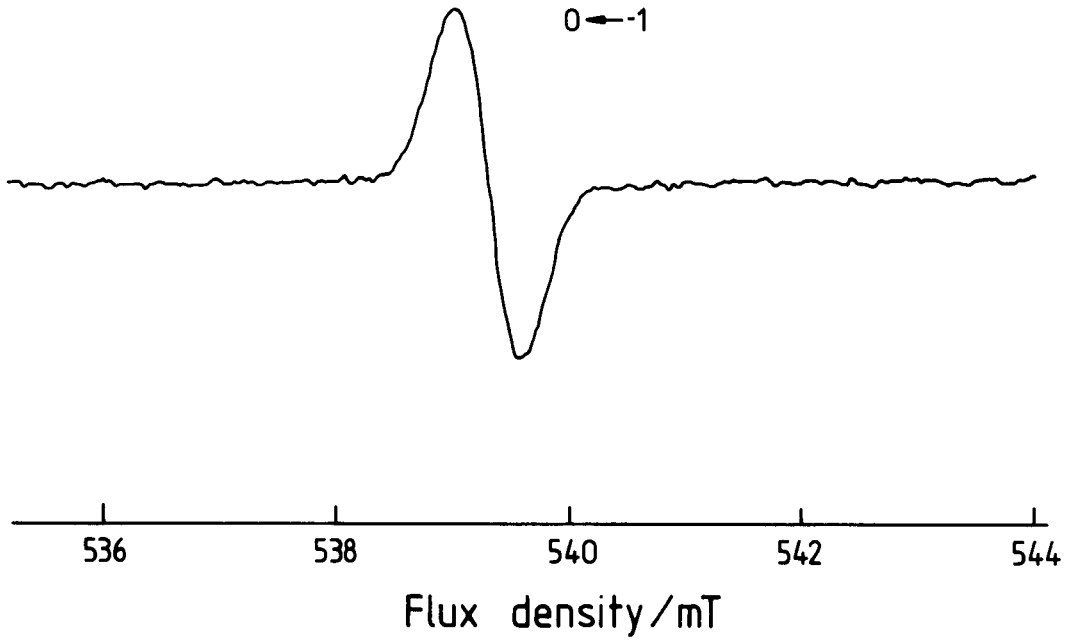

FIG. 2.—Far-infrared laser magnetic resonance spectrum associated with the $J = 0 \leftarrow 1$ transition of Fe in its ground ^5D state, recorded with the 110.7 μm line of CH_3OH, pumped by the 9P(36) line of a CO_2 laser. The spectrum was recorded with the oscillating magnetic field perpendicular to the dc magnetic field ($\Delta M_J = \pm 1$). The transition involved is $M_J = 0 \leftarrow -1$. The output time constant for the lock-in amplifier was 0.3 s.

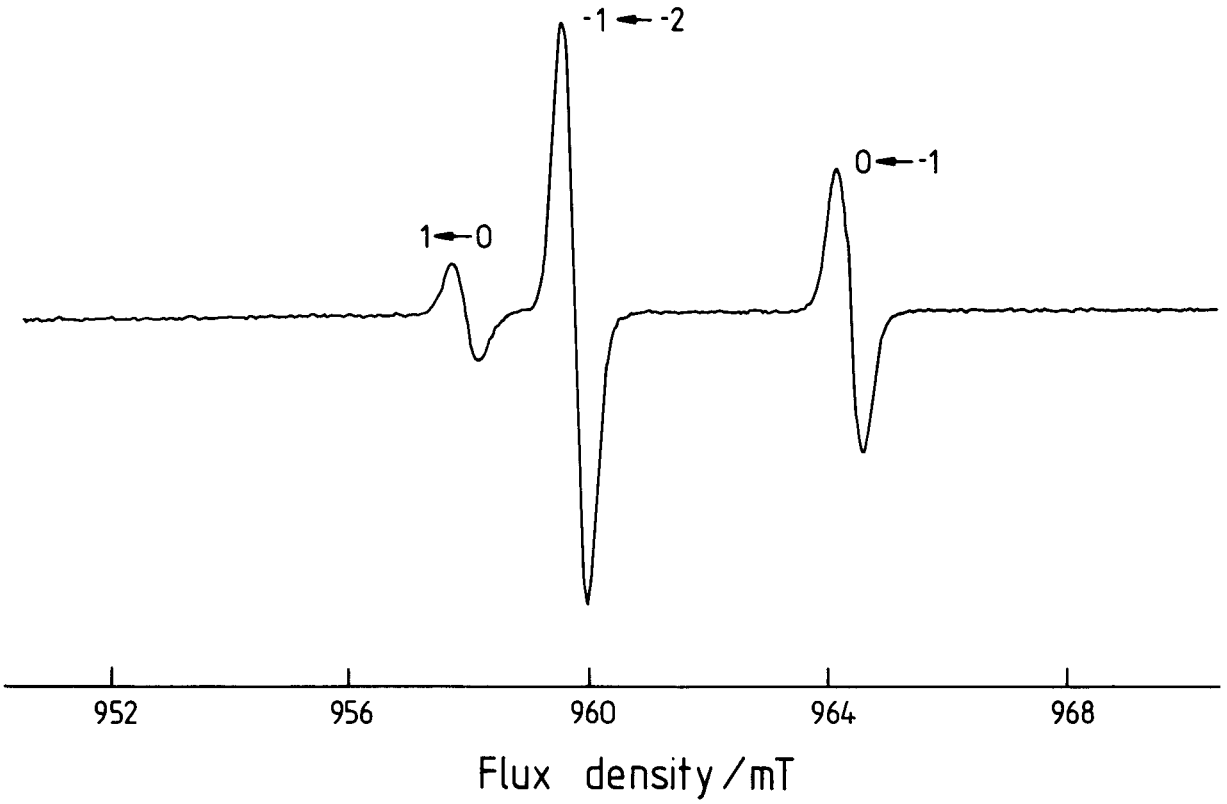

FIG. 3.—Far-infrared laser magnetic resonance spectrum associated with the $J = 1 \leftarrow 2$ transition of Fe in its ground ^5D state, recorded with the 54.1 μm line of CD_3OH, pumped by the 9P(28) line of a CO_2 laser. The spectrum was recorded in perpendicular polarization for magnetic dipole transitions ($\Delta M_J = \pm 1$). The M_J transitions involved are marked in the figure; they have theoretical relative intensities of 1:6:3. The output time constant for the lock-in amplifier was 0.3 s.

TABLE 3

EINSTEIN A- AND B-COEFFICIENTS FOR FINE-STRUCTURE TRANSITIONS IN ATOMIC IRON IN ITS GROUND ^5D STATE

Transition	Frequency (GHz)	$10^4 A_{ij}$ (s^{-1})	$10^{-16} B_{ij}$ (kg^{-1} m)
$^5D_0$–$^5D_1$.....	2696.3942	1.183	2.045
$^5D_1$–$^5D_2$.....	5519.9397	5.920	2.147
$^5D_2$–$^5D_3$.....	8636.13	15.55	1.753
$^5D_3$–$^5D_4$.....	12469.38	25.07	1.022

NOTE.—The A- and B-coefficients have been calculated in the Russell-Saunders limit using the formulae given by Corney 1979.

values, which agree well with our order-of-magnitude more accurate measurements, are given in Table 2 for comparison.

We hope that the accurate measurements of the $J = 0$–1 and $J = 1$–2 fine-structure intervals in Fe 111.2 and 54.3 μm will help the detection of this atom in astrophysical sources. However, it is highly desirable to measure the higher frequency, lower lying fine-structure intervals as well, because they will probably be more useful for astronomical observation. These transitions involve the lower spin components (with a greater population factor), 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. The values for this coefficient for the four fine-structure transitions of atomic iron are also given in Table 3.

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