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# Mössbauer Studies on Iron in the Perovskites $La_{1-x}Sr_xFeO_3$ $(0 \leqslant x \leqslant 1)$†

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Mössbauer spectra of the perovskite series $La_{1-x}Sr_xFeO_y$ $(0.0 \leqslant x \leqslant 1.0;\ y \lesssim 3)$ with $^{57}Fe$ as the photon-emitting and -absorbing nucleus were analyzed. The Néel temperature of antiferromagnetic $LaFeO_3$ decreases with increasing Sr concentration. Characteristic $Fe^{4+}$ lines for high $x$ have isomer shifts between +0.10 and +0.20 mm/sec relative to a source of $^{57}Co$ in stainless steel. However, for intermediate $x$ a $Fe^{3+ \leftrightarrow 4+}$ mixed state seems to exist. Increasing $x$ causes a significant discontinuity in the total isomer shift (transition from magnetic order to disorder). Linewidths vary around 0.40 mm/sec and show broadening in a sample fired briefly.

## INTRODUCTION

The perovskite series $La_{1-x}Sr_xFeO_3$ $(x=0$ to $1)$ was studied by Waugh,[1] and a detailed evaluation of its magnetic properties was undertaken by Kelly.[2] $LaFeO_3$ is antiferromagnetic, with a Néel temperature of about 750°K and weak parasitic ferromagnetism.[3] Substitution of trivalent La by divalent Sr atoms forces the Fe atoms from the stable trivalent to the rare tetravalent state, thereby weakening and even destroying the magnetic order. The present work was undertaken with the expectation that Mössbauer spectroscopy,[4] using $^{57}Fe$ as the $\gamma$-emitting and -absorbing nuclei, might yield new insight.

## PREVIOUS INFORMATION ON THE $La_{1-x}Sr_xFeO_y$ SYSTEM

### Chemical Composition

Oxygen deficiency in the series $La_{1-x}Sr_xFeO_y$ occurs especially at high Sr concentrations and the samples measured by Waugh and Kelly were nonstoichiometric, with 2.7 to 3.0 oxygen ions per formula-molecule, depending on the oxygen pressure during firing. A sample fired at 1300°C *in vacuo* had the formula $SrFeO_{2.5}$ (all iron trivalent)[5]; one of our samples fired for 24 h in 65 atm of oxygen at about 1000°C (no prefiring) had the composition $SrFeO_{2.98}$. While our work was in progress, stoichiometric $SrFeO_3$ has been synthesized[6] by firing at 760°C in 335 atm of oxygen for 7 days.

### Structure

The stoichiometric compounds $La_{1-x}Sr_xFeO_3$ have distorted perovskite structures. $LaFeO_3$ has orthorhombic symmetry with four distorted pseudo-cells in the space group Pbnm, but the structure becomes rhombohedral for the composition $La_{0.5}Sr_{0.5}FeO_3$. With still more Sr, the structure approaches cubic symmetry, and stoichiometric $SrFeO_3$ is cubic.

The compound $SrFeO_{2.5}$ (like $CaFeO_{2.5}$) has the brownmillerite structure[6]: layers of oxygen tetrahedra alternate with layers of oxygen octahedra[7]; the Fe atoms occupy the center sites in both types of polyhedra.

The perovskite $SrFeO_3$ consists of layers of oxygen octahedra. Thus, the $Fe^{4+}$ in $SrFeO_3$ has a uniform octahedron site, while the $Fe^{3+}$ in $SrFeO_{2.5}$ is distributed equally between tetrahedral and octahedral sites.

### Magnetic Properties

According to neutron-diffraction measurements,[8] $LaFeO_3$ has a *G*-type spin-ordered antiferromagnetic structure: Each Fe atom has its spin antiparallel to those of its six nearest Fe neighbors. The Néel temperature of $LaFeO_3$ (750°K) drops as more Sr is introduced.[2] The antiferromagnetism is accompanied by parasitic ferro- or ferrimagnetism as well as by permanent magnetization when $x$ is kept small.[2,5]

For $x \gtrsim 0.4$, all samples proved paramagnetic at room temperature. For $x \rightarrow 1.0$, the Néel temperature drops below 100°K (for $y=2.84$), and the susceptibility $\chi$ increases.

### Mössbauer Measurements

A Mössbauer spectrum identifying the $Fe^{4+}$ line was first reported for $SrFeO_{2.84}$ by Shirane, Cox, and Ruby,[9] who used a source of $^{57}Co$ diffused in type-321 stainless steel. They fitted the experimental points by superposing two pseudo-Lorentzian curves with peaks at

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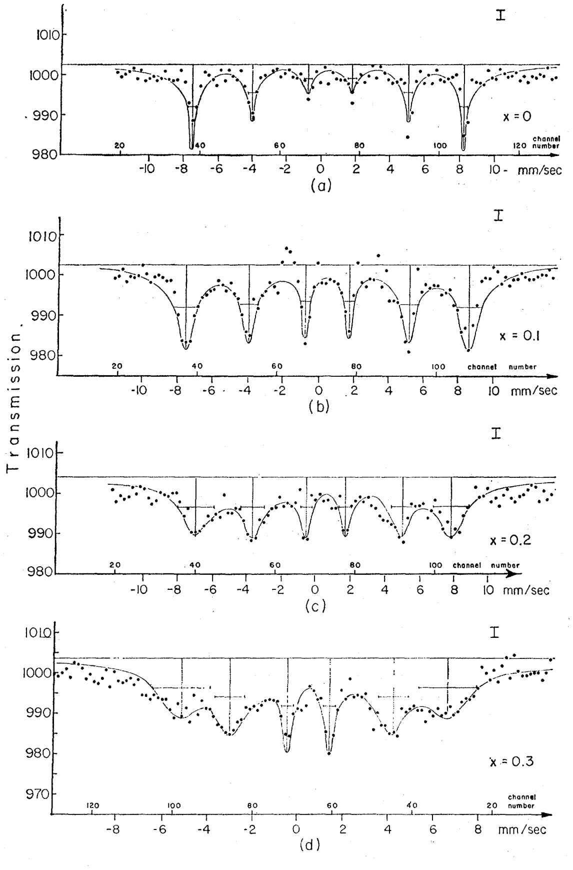

FIG. 1. Spectra of $La_{1-x}Sr_xFeO_3$; $0 \leqslant x \leqslant 0.3$ at (295±5)°K.

velocities 0.10 and 0.55 mm/sec (attributed to $Fe^{4+}$ and $Fe^{3+}$) of relative intensities 3:1 and equal widths (0.35 mm/sec). The relative amounts of $Fe^{4+}$ and $Fe^{3+}$ according to the formula are 68% and 32%, respectively. While this work was in progress, Gallagher *et al.*[6] measured the spectra of $SrFeO_y$ ($y=3.0$, 2.86, 2.60, and 2.5) between 4° and 300°K using a source of $^{57}Co$ in copper. They found for $Fe^{4+}$ a single line at 300°K with an isomer shift of $-0.171$ mm/sec[10] which is split into six lines at and below 78°K; at 4°K it showed an isomer shift of $-0.079$ mm/sec. The sample with $y=2.86$ was found to be a single phase; at 300°K its Mössbauer spectrum showed predominant $Fe^{4+}$ line and magnetic splitting occurred between 4° and 78°K. The sample with $y=2.5$ was resolved as a superposition of tetrahedral and octahedral sites.

TABLE I. Parameters of room-temperature spectra of $La_{1-x}FeO_3$; $0 \leqslant x \leqslant 0.3$.

| | $x=0.0$ [Fig. 1(a)] | $x=0.1$ [Fig. 1(b)] | $x=0.2$ [Fig. 1(c)] | $x=0.3$ [Fig. 1(d)] |
|---|---|---|---|---|
| $\epsilon_{is}$ (mm/sec) | 0.432 | 0.525 | 0.566 | 0.580 |
| $H_{int}$ (kOe) | 480 | 499 | 476 | 384 |
| $\frac{Fe^{4+}}{\text{total Fe}}$ (%) (chem. anal.) | 0.5 | 9.8 | 20.0 | 32.8 |

TABLE II. Room-temperature parameters of $La_{1-x}Sr_xFeO_y$, $0.4 \leqslant x \leqslant 0.7$.

| | $x=0.4$ | $x=0.5$ | $x=0.6$ | $x=0.7$ |
|---|---|---|---|---|
| $\eta$ | 0.0861 | 0.191 | 0.48 | 0.194 |
| Γ (mm/sec) | 0.573 | 0.432 | 0.425 | 0.465 |
| $\epsilon_{is}$ (mm/sec) | 0.312 | 0.308 | 0.274 | 0.247 |
| $\frac{Fe^{4+}}{\text{total Fe}}$ (%) (chem. anal.) | 34.8 | 50 | 54 | 62 |
| $y$ | 2.97 | 3.00 | 2.97 | 2.96 |

## EXPERIMENTAL

### Equipment and Samples

The radioactive source in the Mössbauer spectrometer was driven by an electromechanical system providing a triangular velocity wave form.[11] The detector of the γ photons was an argon-methane proportional counter with a thin Lucite window. After rejection of background noise, the pulses corresponding to the $^{57}Fe$ 14.4-eV photons were accumulated in a 256-channel analyzer which sorted them according to the velocity of the source.

Velocity calibration of the curves was based on the positions of the six absorption lines of a $^{57}Fe$-enriched standard $Fe_2O_3$ absorber, measured with an accuracy of three decimal places.[12] Their values are $-7.89$, $-4.17$, $-0.76$, $+1.92$, $+5.36$, and $+8.59$ mm/sec, relative to a source of $^{57}Co$ in type-310 stainless steel. Before and after each run the spectrum of the absorber served to calibrate the horizontal channel axis. The parameters given in the tables are accurate to ca. ±5%.

Samples were prepared by pulverizing the compounds, taking the amount necessary for ca. 37% ($=1/e$) nonresonant transmission, mixing it uniformly with hot Carbo-Wax 4000 (Union Carbide Corp.), and then casting a disk (diam 23 mm, thickness 3 mm).

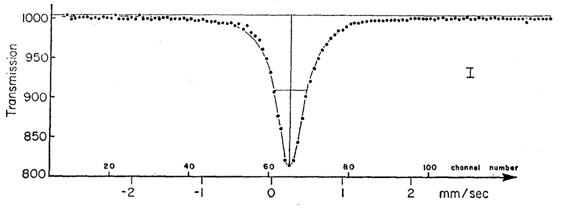

FIG. 2. Spectrum of $La_{1-x}Sr_xFeO_3$; $x=0.7$ at (295±5)°K.

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[10] The difference in the $Fe^{4+}$ isomer shifts reported above results from the different hosts for the $^{57}Co$ source.

[11] R. L. Cohen, P. G. McMullin, and G. K. Wertheim, Rev. Sci. Instr. 34, 671 (1963).

[12] O. C. Kistner and A. W. Sunyar, Phys. Rev. Letters 4, 412 (1960).

TABLE III. Room-temperature parameters of $La_{1-x}Sr_xFeO_y$, $x=0.8$, 0.9.

| | | $x=0.8$ | $x=0.9$ |
|---|---|---|---|
| Left line | $\eta$ | 0.21 | 0.253 |
| | $\Gamma$ (mm/sec) | 0.55 | 0.499 |
| | $\xi$ (mm/sec) | +0.15 | +0.145 |
| Right line | $\eta$ | 0.057 | 0.053 |
| | $\Gamma$ (mm/sec) | 0.38 | 0.294 |
| | $\xi$ (mm/sec) | +0.70 | +0.680 |
| $\frac{Fe^{4+}}{\text{total Fe}}$ (%) (chem. anal.) | | 60.9 | 73.5 |
| $y$ | | 2.90 | 2.92 |

They were chemically analyzed[13] for $Fe^{4+}$ in reference to the total amount of Fe; from this the oxygen content was calculated. The results were found to scatter appreciably. Most samples for the system $La_{1-x}Sr_xFeO_3$ were those used by Waugh and Kelly in their measurements. R. L. Anderson of the Ceramics Department at MIT also provided several samples. At low temperatures, measurements were performed with the sample in a Dewar and the source outside. The sample temperature fluctuated within ca. ±5°K.

## EXPERIMENTAL RESULTS

### Mössbauer Spectra and Parameters

In the Mössbauer spectra shown the horizontal axis is the velocity axis calibrated in mm/sec and in channel numbers. The vertical scale gives the transmission of the absorber in relative counts per unit velocity, $dn/dv$; this scale is chosen so that 1000 corresponds approximately to the value of transmission far away from resonance $[(dn/dv)_{v\to\infty}]$. In the right-hand side the statistical standard deviation for the recorded points caused by the radioactive decay is indicated. The actual spread of experimental points also include statistical uncertainties of the driving and recording systems.

The points shown are experimental, and the solid line is a calculated superposition of Lorentzian functions. The base line of the latter $[(dn/dv)_{v\to\infty}]$ and the three parameters for each component peak (velocity of peak $\xi$, intensity $\eta$, and width $\Gamma$) were found by fitting a least-squares Lorentzian curve to the experimental points by means of a computer program based on the method of "variable metric minimization."[14]

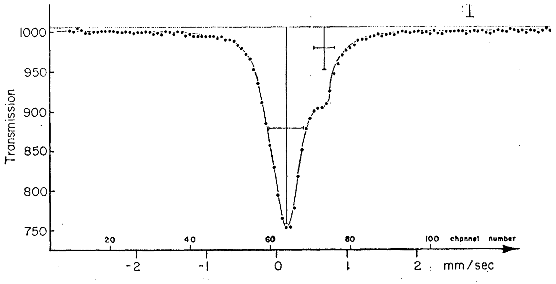

FIG. 3. Spectrum of $La_{1-x}Sr_xFeO_{2.92}$; $x=0.9$ at $(295\pm5)$°K.

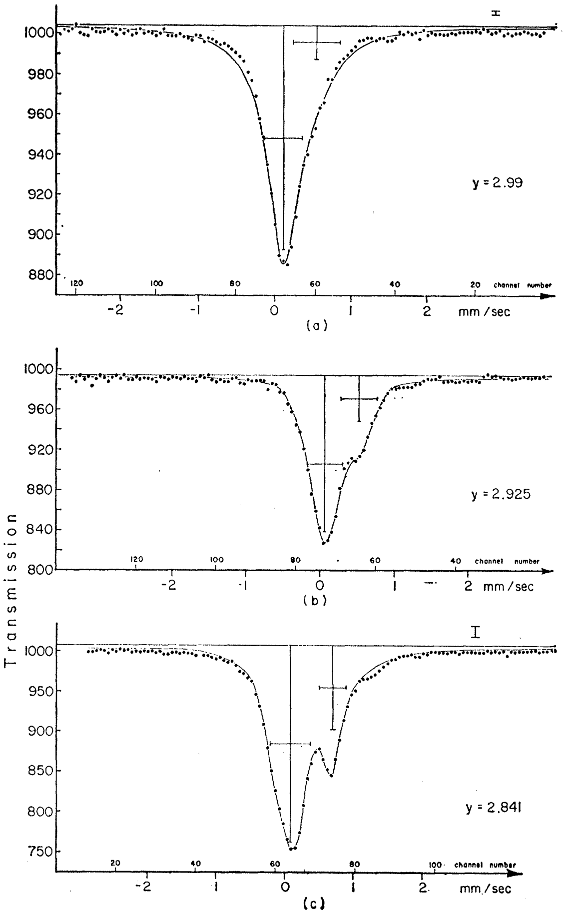

FIG. 4. Spectra of $SrFeO_y$ at $(295\pm5)$°K.

TABLE IV. Room-temperature parameters of paramagnetic $SrFeO_y$.

| | | $y=2.99$ | $y=2.925$ | $y=2.841$ |
|---|---|---|---|---|
| Left line | $\eta$ | 0.112 | 0.166 | 0.245 |
| | $\Gamma$ (mm/sec) | 0.519 | 0.510 | 0.588 |
| | $\xi$ (mm/sec) | 0.142 | 0.070 | 0.097 |
| Right line | $\eta$ | 0.016 | 0.065 | 0.105 |
| | $\Gamma$ (mm/sec) | 0.618 | 0.550 | 0.390 |
| | $\xi$ (mm/sec) | 0.511 | 0.520 | 0.706 |
| $\frac{Fe^{4+}}{\text{total Fe}}$ (%) (chem. anal.) | | 98 | 85 | 68.2 |
| $\epsilon_{is}$ | | 0.193 | 0.205 | 0.253 |

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[13] By D. L. Guernsey and T. F. Strow, Analytical Laboratory, Metallurgy Department, MIT, Cambridge, Massachusetts.

[14] W. C. Davidon, Argonne National Laboratory Report No. ANL-5990 (unpublished).

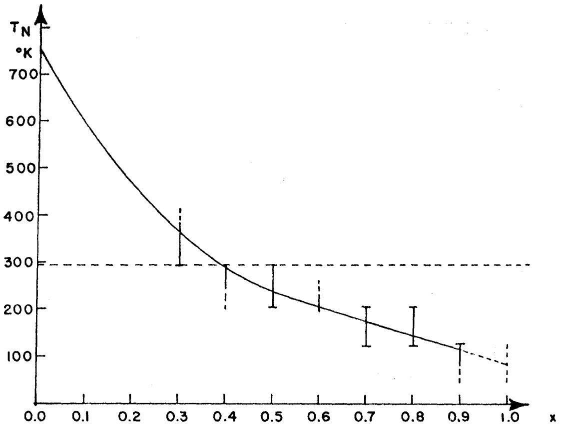

FIG. 5. Néel temperatures of $La_{1-x}Sr_xFeO_3$.

### Room-Temperature Spectra of $La_{1-x}Sr_xFeO_y$ for $0.0 \leqslant x \leqslant 0.3$

At room temperature, the Mössbauer spectra of the four compounds discussed here ($x=0.0$, 0.1, 0.2, 0.3) show the six lines characteristic of the nuclear Zeeman effect (Fig. 1). These compounds are antiferromagnetic at room temperature, and the local field at the nuclei is very high ($H_{\text{int}} \simeq 450$ kOe) despite low bulk magnetization due to opposite orientation of neighboring Fe spins. The best resolved lines are those of $LaFeO_3$ [Fig. 1(a)]; since the bandwidths of the lines are almost equal, a Lorentzian fit could be found with the intensity ratios of 3:2:1:1:2:3, although the spectrum shows some saturation. As $x$ increases, the external lines tend to broaden and decrease in intensity. Still, the area ratios of the lines can be approximated 3:2:1:1:2:3. The rather blurred lines at $x=0.3$ [Fig. 1(d)] suggest that its Néel temperature is only slightly above room temperature. Table I summarizes the isomer shift as well as the internal calculated magnetic field at the Fe nuclei $H_{\text{int}}$, and the fraction of $Fe^{4+}$, based on the chemical analysis.

### Room-Temperature Spectra of $La_{1-x}Sr_xFeO_y$, $0.4 \leqslant x \leqslant 1.0$

As more Sr is substituted for La, a dramatic change occurs in the room-temperature spectra somewhere between $x=0.3$ and 0.4. The six-line spectrum still recognizable at $x=0.3$ coalesces into a single-line spectrum at $x=0.4$, due to the transition from antiferromagnetic (or ferrimagnetic) to paramagnetic behavior in which both the local magnetic field and the nuclear Zeeman splitting vanish. The parameters are summarized in Table II and a typical spectrum is shown in Fig. 2.

TABLE V. Low-temperature parameters of $La_{1-x}Sr_xFeO_3$ spectra with hyperfine splitting.

| | Coolant—dry ice | | Coolant—liquid N | | |
|---|---|---|---|---|---|
| | $x=0.3$ | $x=0.4$ | $x=0.4$ | $x=0.5$ | $x=0.6$ |
| $T$ (°K) | 218 | 207 | 152 | 109 | 124 |
| $\epsilon_{is}$ (mm/sec) | 0.460 | 0.741 | 0.165 | 0.39 | 0.79 |
| $H_{\text{int}}$ (kOe) | 452 | 386 | 491 | 480 | 443 |

TABLE VI. Low-temperature parameters of paramagnetic $La_{1-x}Sr_xFeO_y$ spectra.

| | $x=0.7$<br>$y=2.96$ | $x=0.8$<br>$y=2.90$ | $x=1.0$<br>$y=2.84$ |
|---|---|---|---|
| $T$ (°K) | 210 | 128 | 88 |
| $\eta$ | 0.33 | $\begin{cases}0.054\\0.016\end{cases}$ | 0.086<br>0.021 |
| $\Gamma$ (mm/sec) | 0.54 | $\begin{cases}1.66\\1.35\end{cases}$ | 1.10<br>0.79 |
| $\xi$ (mm/sec) | 0.53 | $\begin{cases}0.72\\1.82\end{cases}$ | −0.37<br>+0.40 |
| $\epsilon_{is}$ (mm/sec) | 0.53 | 0.93 | +0.161 |

Samples with a Sr concentration of $x=0.8$, 0.9 display small peaks on the positive-velocity slope of the prominent line (Fig. 3 and Table III).

Figure 4 shows $SrFeO_y$ ($x=1$) spectra for three values of $y$, and Table IV the corresponding parameters and values of the $Fe^{4+}$ fraction and of $y$. The dominant single peak at $y=3.0$ develops a small peak on its positive-velocity slope, which grows as $y$ decreases. The various $y$ values were achieved by varying the oxygen pressure under which the samples were fired, as well as the temperatures and prefiring routines.

### Low-Temperature Measurements of the Series $La_{1-x}Sr_xFeO_y$

Low-temperature spectra were recorded only for compounds that are paramagnetic at room temperature, to find the temperature range in which magnetic ordering appears. Fe impurity in the Be windows of the Dewar chamber introduced an absorption line which had to be removed.

Spectra taken with dry ice as a coolant (samples at ca. 210°K) show a magnetic splitting for $x=0.5$; for

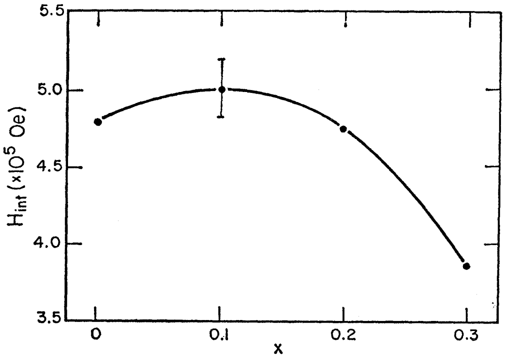

FIG. 6. Room-temperature quadrupole splitting $E_Q$ and internal magnetic field $H_{\text{int}}$ of $La_{1-x}Sr_xFeO_3$, $0 \leqslant x \leqslant 0$, 3.

$x=0.6$ a single line appears, indicating paramagnetism. At $x=0.5$ there is already considerable blurring of the lines, suggesting that the transition temperature is slightly above 210°K. The spectra taken with liquid nitrogen as coolant (samples at ca. 125°K) indicate a magnetic splitting up to $x=0.7$, which disappears for $x \geqslant 0.8$. A spectrum of $SrFeO_{2.84}$ with liquid helium as coolant (sample at 16°K) shows incipient magnetic splitting. Parameters of the magnetically split spectra, which allow resolution of the lines, are listed in Table V, and those showing paramagnetic behavior in Table VI.

## DISCUSSION

### Magnetic Properties

The Mössbauer spectra demonstrate that substitution of Sr for La lowers the Néel Temperature (Fig. 5).

The internal magnetic field $H_{\text{int}}$ as function of $x$ at room temperature shows a maximum near $x=0.1$ and a decrease to zero for $0.3<x<0.4$ (Fig. 6). Dry-ice and liquid-nitrogen measurements show a decrease in $H_{\text{int}}$ as $x$ increases (cf. Table V), as anticipated.

### Trivalent and Tetravalent Iron States

Isomer shifts in iron compounds indicate a correlation between the isomer shift and the valency state of the iron (Fig. 7).[15] The $SrFeO_y$ spectra (cf. Fig. 4) and those of $La_{0.1}Sr_{0.9}FeO_y$ and $La_{0.2}SrFeO_y$ (cf. Fig. 3) show a dominant peak at 0.10 to 0.15 mm/sec, and a smaller peak (or peaks) at about 0.50 to 0.70 mm/sec. Since the latter falls in or close to the range of ionic $Fe^{3+}$, it is natural to conclude[6,9] that the dominant peak corresponds to the $Fe^{4+}$ state and to allocate the region of between 0.10 and 0.15 mm/sec to this state. Theoretical calculations[16] for the isomer shift of *free ionic* $Fe^{4+}$ predict a value of about $-1.30$ mm/sec, much too negative compared to the experimental results with *chemically bound* $Fe^{4+}$. It seems that the bound $Fe^{4+}$ ions are more shielded from the nucleus, and the isomer shift becomes more positive than that predicted for free $Fe^{4+}$.

### Evidence for an Intermediate Valency between $Fe^{3+}$ and $Fe^{4+}$

An attempt to find further evidence for the presence of *distinct* $Fe^{4+}$ and $Fe^{3+}$ lines in the spectra of $La_{1-x}Sr_xFeO_y$, $x=0.4$, 0.5, 0.6, and 0.7 failed (cf. Fig. 2). Here in spite of an appreciable proportion of $Fe^{4+}$ (cf. Table II), no splitting into $Fe^{3+}$, $Fe^{4+}$ lines is noticeable. The single narrow line in these spectra has an isomer shift somewhere between the $Fe^{3+}$ and $Fe^{4+}$ regions. The true oxidation state seems to be neither $Fe^{3+}$ nor $Fe^{4+}$ but an "intermediate" state. The rhombohedral symmetry[1] of the compounds with $0.4<x<0.7$ might be associated with this state and provides the proper space geometry.

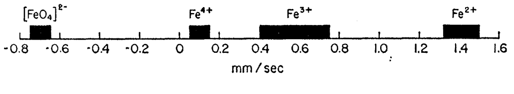

FIG. 7. Isomer shifts of iron compounds referred to a source of $^{57}Co$ in stainless steel 310.

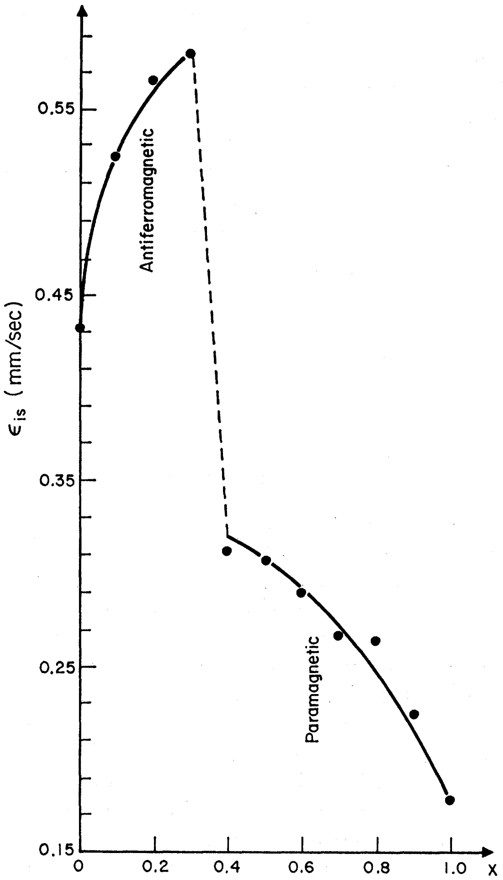

FIG. 8. Isomer shift versus Sr concentration.

### Isomer Shift as a Function of Sr Concentration

The room-temperature isomer shifts $\epsilon_{is}$ of the series $La_{1-x}Sr_xFeO_y$ (Fig. 8) show that in the region where magnetic splitting occurs ($0.0 \leqslant x \leqslant 0.3$), $\epsilon_{is}$ tends to increase with $x$ until transition to paramagnetic behavior takes place ($0.3<x<0.4$). Here it drops drastically and then decreases with increasing $x$. Despite the fact that the velocity scales and methods of finding $\epsilon_{is}$ differ for the magnetically split and the paramagnetic spectra, this sudden change in the isomer shift in the transition from magnetic order to paramagnetic appears to be real.

A similar though smaller discontinuity in the

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[15] E. Fluck, W. Kerler, and W. Neuwirth, Angew. Chem. Intern. Ed. Engl. **2**, 277 (1963).

[16] L. R. Walker, G. K. Wertheim, and V. Jaccarino, Phys. Rev. Letters **6**, 98 (1961).

isomer shift of metallic iron in transition through its Curie temperature has been reported,[17] and several mechanisms to explain it have been discussed in the literature.[17–19]

## ACKNOWLEDGMENTS

The authors wish to express their thanks to Professor A. R. von Hippel for suggesting the problem and for helpful discussions, and to Professor L. Grodzins and Dr. R. Bauminger for assistance and use of equipment. Computer time was donated by the Computation Center, MIT.

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[19] S. Alexander and D. Treves, The Weizmann Institute of Science, Department of Electronics, Rehovoth, Israel, Technical Note No. 16, 1965 (unpublished).

# Errata

**Theory of Resonance Broadening of Spectral Lines by Atom-Atom Impacts,** A. W. ALI AND H. R. GRIEM [Phys. Rev. **140**, A1044 (1965)]. In taking averages over the directions of the vectors $\boldsymbol{\rho}$ and $\mathbf{v}$, correlations between their components due to orthogonality and normalization of these vectors were treated incorrectly. With corrected relations for the averages like $\{\rho_x^2\rho_y^2\}=(1/15)\rho^4$, $\{\rho_x^2v_x^2\}=(1/15)\rho^2v^2$, and $\{\rho_x\rho_yv_xv_y\}=-\frac{1}{2}\{\rho_x^2v_x^2\}$, the coefficients in Eq. (7) become $+2/5$ and $+2/15$ instead of $+10/9$ and $-2/9$. In all subsequent relations for the second-order term, the correct numerical coefficient is obtained by substituting "3" for "7" (or "9" for "21"). Using especially the orthogonality relation to re-evaluate the fourth-order term, the quantity $\epsilon$ is now calculated to be exactly 0.25 rather than 0.62. The numerical factor in the final result is thus $2(1-1/24)\approx 1.92$ as compared to 2.74 in Eq. (22) of the original paper. This new resonance width is only 6% larger than Byron and Foley's result (Ref. 4). Note also that the paper by Tsao and Curnette (Ref. 5) has been published in J. Quant. Spectr. Radiative Transfer **2**, 41 (1962).

**Theory of Wing Broadening of the Hydrogen Lyman-α Line by Electrons and Ions in a Plasma,** HANS R. GRIEM [Phys. Rev. **140**, A1140 (1965)]. As recognized by G. Peach, Eq. (10) represents a binomial series multiplied with $\frac{9}{2}(u_x^2+u_y^2)$, the series yielding $3^{2(l-1)}$. The right-hand side of Eq. (11) thus becomes $3^{2l-1}$. This differs from the original result because of the incorrect use there, e.g., of $\{u_x^2u_y^2\}=\frac{1}{9}$ rather than of $\{u_x^2u_y^2\}=1/15$. To correct Eq. (12), the triple sum (multiplied with $\frac{3}{2}$) must be replaced by

$$\sum_{l=2}^{\infty}\frac{(-1)^l}{(l-1)(2l)!}\left(\frac{6\hbar}{m\rho v}\right)^{2(l-1)},$$

resulting in new constants or coefficients of $-0.891_5$ and $\frac{3}{2}$ for Eq. (14) rather than $-0.967$ and 19/10. Using relations like $\{\rho_x^2\rho_y^2\}=(1/15)\rho^4$, $\{\rho_x^2v_x^2+2\rho_x\rho_yv_xv_y\}=0$, and $\{\rho_x^2v_x^2+2\rho_x^2v_y^2\}=\frac{1}{3}\rho^2v^2$, the coefficient of the quadrupole correction term in Eq. (17) is now calculated as 8 instead of 28. In the final results, i.e., in Eqs. (32b) and (34), the coefficient 14/3 must therefore be replaced by $\frac{4}{3}$ and the constant term $\frac{1}{10}$ be doubled, because in the original calculations a constant term $-1$ was used instead of the correct value $-0.891_5$. For the plasma conditions and wavelength ranges of the arc (Ref. 10) and shock tube (Ref. 9) experiments, these corrections result in reductions of the intensities by at most 5 and 7%, respectively.