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NONMAGNETIC IMPURITIES

Jag J. Singh

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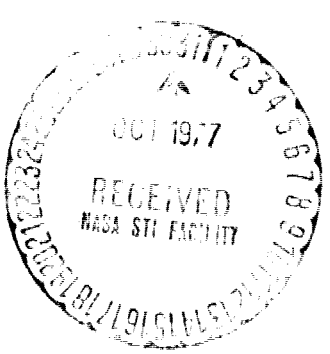

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ABSTRACT

We have recently completed a study of Mossbauer parameters of dilute quaternary (Fe-XYZ) alloys where X, Y, Z, are Al, Ge, and La, respectively. A comparison between the computed and the observed values indicates that changes in effective hyperfine field (-0.37 $\pm$ 0.04% per a/o), outer peak widths (+16.30 $\pm$ 0.65% per a/o) and the isomer shift (-0.90 $\pm$ 0.04% per a/o) predicted on the basis of binary alloys of the individual impurities are equal - within experimental errors - to those obtained in quaternary (Fe-XYZ) alloys. This result indicates that the effects of simultaneously present dilute impurities are additive, independent of their respective electronic configurations.

INTRODUCTION

As a part of continuing research[1] to extend the applicability of Mossbauer effect to the study of defects and their diffusion in complex metallic systems, we recently completed[2] an investigation of the dilute Fe-X alloys where X was Al, Ge, or La. Two of the solute atoms (Al, Ge) are part of the s-p series whereas one (La) is a part of the s-d series. In view of the reported dissimilarity[3] of the hyperfine field patterns at the solute atoms of these two series, it was decided to measure the Mossbauer

parameters of dilute (Fe-XYZ) alloys where X, Y, Z are Al, Ge, and La, respectively. An additional motivation behind this extension was to confirm that the effects of simultaneously present impurities at low concentration are additive. Such studies are very relevant to understanding the effects of simultaneous presence of various types of impurity atoms and defects in the critical regions of the fatigue test structures.

EXPERIMENTAL PROCEDURE

The (Fe-XYZ) alloys were prepared by induction melting of the mixture of four elements in an argon atmosphere in an alumina crucible. The solute concentrations were varied from 0.5 to 2.0 atomic percent (a/o), in steps of 0.5 a/o, for each dilute constituent. The alloy specimens were cold-rolled into foils, 0.001-inch thick, and cut into 1" x 1" pieces for use as Mossbauer targets.

A conventional electromagnetic drive system[4] moved a 25 mc Co^{57} (Pt) source at a constant acceleration to provide transmission Mossbauer spectra of these targets. An examination of these spectra indicated that there were no evident shoulders to the hyperfine peaks, indicating the validity of the concept of effective hyperfine field at iron nuclei in dilute alloys. These spectra were computer analyzed[5] to determine the following parameters: (1) Effective hyperfine field at the iron nucleus, (2) full width at half maximum height (FWHM) for peaks 1 and 6, and (3) isomer shift.

In addition, the binary alloy study, which was previously limited[2] to 2 a/o solute concentration, was extended to 5 a/o concentration in order to determine more accurate values of rates of changes of the three Mossbauer

parameters with the dilute constituent concentrations. These rate parameters are needed to calculate the corresponding rate parameters for the quaternary Fe-XYZ alloys.

The results are discussed in the following paragraphs.

RESULTS AND DISCUSSION

Figure 1 shows two binary alloy spectra for solute concentrations of 5 a/o. Figure 2 shows Fe-XYZ alloy spectrum at 2 a/o concentration of each of the dilute constituents. A pure iron spectrum is also shown in these figures for comparison purposes. It is obvious that the alloy spectra look similar to the single component pure iron hyperfine spectrum and can therefore be fitted with a single Lorentzian line to obtain the values of effective Mossbauer parameters. The values of effective hyperfine field (H_{eff}), effective width (Γ_{eff}), and effective isomer shift ($I.S._{eff}$) thus obtained for the three binary alloys for several values of impurity concentrations are summarized in Tables I, II, and III. Figures 3-5 illustrate these results. Mossbauer parameters for (Fe-XYZ) alloys are summarized in Tables IV-VI. (Also included in these tables are the values computed on the basis of the data for binary Fe-X alloys of corresponding total impurity level.) The computed values have been obtained as follows:

$$H_{eff}(\text{Fe-XYZ}) = \frac{1}{3}\left[H_{eff}(\text{Fe-X}) + H_{eff}(\text{Fe-Y}) + H_{eff}(\text{Fe-Z})\right] \qquad (1)^{(*)}$$

(*)Magnetic fields (H_{eff} and H_{Fe}) have been calculated as follows:

$$H = 1/3(2H_o + H_1) \qquad (1a)$$

where H_o = magnetic field based on Fe^{57} ground state splitting
H_1 = magnetic field based on Fe^{57} first excited state splitting.

$$\Gamma_{eff}(Fe\text{-}XYZ) = \left[\frac{1}{3}\left\{\Gamma_{eff}^{2}(Fe\text{-}X) + \Gamma_{eff}^{2}(Fe\text{-}Y) + \Gamma_{eff}^{2}(Fe\text{-}Z)\right\}\right]^{1/2} \quad (2)$$

$$I.S._{eff}(Fe\text{-}XYZ) = \frac{1}{3}\left[I.S._{eff}(Fe\text{-}X) + I.S._{eff}(Fe\text{-}Y) + I.S._{eff}(Fe\text{-}Z)\right] \quad (3)$$

Data summarized in Tables IV-VI are illustrated in figures 6, 7, and 8. The solid lines in these figures are the best fit through the computed values. It is apparent that the observed and the computed values of the three parameters are in agreement, within experimental errors.

From the data summarized in Tables IV-VI and illustrated in figures 6, 7, and 8, the following values of changes in the three parameters, with the impurity concentrations, have been obtained for the (Fe-XYZ) alloys:

$$\Delta H_{eff} = -(0.37 \pm 0.04\%) \text{ per a/o} \quad (4)$$

$$\Delta \Gamma_{eff} = +(16.30 \pm 0.65\%) \text{ per a/o} \quad (5)$$

$$\Delta I.S._{eff} = -(0.90 \pm 0.04\%) \text{ per a/o.} \quad (6)$$

From the above results, it is apparent that the effects of dilute constituents in solid solution in iron are not affected by the simultaneous presence of other low concentration solutes. This is an extension of the previously reported[6] additive law for the effects of identical impurity atoms in various neighbor shells around the iron atoms. Two reasons can be postulated for the observed behavior in the present study: (1) There is small likelihood of different low concentration solute atoms interacting with

each other and thereby affecting their individual interactions with the iron atoms; (2) the Al($3p^1$ $3s^2$) and Ge($4p^2$ $4s^2$) atoms affect the 3d electron population in iron in similar ways whereas La($5d^1$ $6s^2$) atoms barely affect the iron structure. However, the extended additive law should hold for all low concentration solute atoms, regardless of their electronic structure, as long as no clustering of the impurity atoms - which can affect their individual effects as well as upset the random distribution of the alloy atoms - takes place.

CONCLUDING REMARKS

From the results discussed above, it would appear that the effects of simultaneously present dilute impurities are additive, independent of their respective electronic configurations. This result is significant since it allows computation of the Mossbauer parameters of impure specimens without having to resolve the experimental spectra into individual constituent spectra. Furthermore, the concept of "effective" hyperfine field and the related parameters indicates the usefulness of Mossbauer spectrometry for practical structural applications where the targets are often of uncertain composition or alloys containing several low concentration impurities.

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TABLE I.- Summary of H_{eff} values in (Fe-Al), (F-Ge), and (Fe-La) alloys as a function of solute concentration

No.	Solute Concentration a/o	H_{eff}/H_{Fe} Values		
		(Fe-Al)	(Fe-Ge)	(Fe-La)
1	0.01	1.0000	1.0000	1.0000
2	0.5	0.9986 ± 0.0031	0.9983 ± 0.0024	0.9979 ± 0.0019
3	1.0	0.9962 ± 0.0019	0.9951 ± 0.0020	0.9995 ± 0.0032
4	1.5	0.9950 ± 0.0021	0.9906 ± 0.0015	0.9972 ± 0.0020
5	2.0	0.9924 ± 0.0030	0.9906 ± 0.0019	0.9919 ± 0.0060
6	3.0	0.9864 ± 0.0031	0.9934 ± 0.0030	0.9936 ± 0.0052
7	4.0	0.9839 ± 0.0026	0.9857 ± 0.0051	0.9921 ± 0.0039
8	5.0	0.9779 ± 0.0024	0.9852 ± 0.0029	0.9933 ± 0.0029
Rate of change/a/o		-0.44%	-0.36%	-0.19%

TABLE II.- Summary of $(\Gamma_{eff})_{1,6}$ values in (Fe-Al), (Fe-Ge), and (Fe-La) alloys as a function of the solute concentration

No.	Solute Concentration a/o	$\left(\frac{\Gamma_{eff}}{\Gamma_{Fe}}\right)_{1,6}$ Values		
		(Fe-Al)	(Fe-Ge)	(Fe-La)
1	0.0	1.0000	1.0000	1.0000
2	0.5	1.0615 ± 0.0207	1.0356 ± 0.0120	1.1190 ± 0.0200
3	1.0	1.2232 ± 0.0100	1.2212 ± 0.0140	1.2061 ± 0.0200
4	1.5	1.2802 ± 0.0141	1.3403 ± 0.0400	1.0271 ± 0.0166
5	2.0	1.3872 ± 0.0345	1.5000 ± 0.0500	1.1400 ± 0.0500
6	3.0	1.5176 ± 0.0296	1.6248 ± 0.0623	1.1237 ± 0.0442
7	4.0	1.7240 ± 0.0143	1.9394 ± 0.0860	1.2205 ± 0.0509
8	5.0	1.9870 ± 0.0440	2.4209 ± 0.3025	1.1050 ± 0.0486
Rate of change/a/o		+17.3%	+26.0%	+4.3%

TABLE III.- Summary of $(I.S.)_{eff}$ values in (Fe-Al), (Fe-Ge), and (Fe-La) alloys as a function of solute concentration

No.	Solute Concentration	$(I.S.)_{eff}/(I.S.)_{Fe}$ Values		
		(Fe-Al)	(Fe-Ge)	(Fe-La)
1	0.0	1.0000	1.0000	1.0000
2	0.5	0.9992 ± 0.0065	0.9943 ± 0.0062	1.0044 ± 0.0081
3	1.0	0.9984 ± 0.0078	0.9846 ± 0.0078	0.9977 ± 0.0063
4	1.5	0.9937 ± 0.0083	0.9792 ± 0.0100	0.9930 ± 0.0065
5	2.0	0.9904 ± 0.0096	0.9656 ± 0.0128	0.9924 ± 0.0068
6	3.0	0.9769 ± 0.0141	0.9705 ± 0.0116	1.0000 ± 0.0205
7	4.0	0.9828 ± 0.0156	0.9852 ± 0.0194	0.9897 ± 0.0205
8	5.0	0.9661 ± 0.0185	0.9219 ± 0.0228	0.9778 ± 0.0189
Rate of change/a/o		-0.64%	-1.78%	-0.34%

TABLE IV

COMPARISON BETWEEN THE COMPUTED(*) AND THE MEASURED VALUES OF (H_{EFF}/H_{FE}) FOR DILUTE (Fe-AlGeLa) ALLOYS

NO.	DILUTE SOLUTE CONCENTRATION (%)	H_{EFF}/H_{FE} VALUE	
		MEASURED VALUE	COMPUTED VALUE
1	0.5% EACH	0.9976 ± 0.0052	0.9973 ± 0.0021
2	1.0% "	0.9909 ± 0.0063	0.9906 ± 0.0032
3	1.5% "	0.9813 ± 0.0063	0.9855 ± 0.0025
4	2.0% "	0.9742 ± 0.0064	0.9802 ± 0.0039

(*) COMPUTED $H_{EFF} = \frac{1}{3}\left[H_{EFF}(Fe\text{-}Al) + H_{EFF}(Fe\text{-}Ge) + H_{EFF}(Fe\text{-}La)\right]$

TABLE V

COMPARISON BETWEEN THE COMPUTED(*) AND THE MEASURED VALUES OF (Γ/Γ_{Fe}) FOR DILUTE (Fe-AlGeLa) ALLOYS

NO.	DILUTE SOLUTE CONCENTRATION (%)	Γ/Γ_{Fe} VALUES	
		MEASURED VALUE	COMPUTED VALUE
1	0.5% EACH	1.1179 ± 0.0920	1.2237 ± 0.0250
2	1.0% "	1.6482 ± 0.1625	1.4400 ± 0.0510
3	1.5% "	1.8562 ± 0.1607	1.7557 ± 0.0720
4	2.0% "	1.9220 ± 0.1834	1.9590 ± 0.1830

(*) COMPUTED $\Gamma = \left[\frac{1}{3}\left\{\Gamma^2(Fe\text{-}Al) + \Gamma^2(Fe\text{-}Ge) + \Gamma^2(Fe\text{-}La)\right\}\right]^{1/2}$

TABLE VI

COMPARISON BETWEEN THE COMPUTED(*) AND THE MEASURED VALUES OF $(I.S./I.S.)_{Fe}$ FOR DILUTE (Fe-AlGeLa) ALLOYS

NO.	DILUTE SOLUTE CONCENTRATION (%)	I.S./$(I.S.)_{Fe}$ VALUE	
		MEASURED VALUE	COMPUTED VALUE
1	0.5% EACH	0.9953 ± 0.0155	0.9866 ± 0.0083
2	1.0% "	0.9639 ± 0.0191	0.9724 ± 0.0081
3	1.5% "	0.9772 ± 0.0215	0.9586 ± 0.0094
4	2.0% "	0.9560 ± 0.0243	0.9448 ± 0.0101

(*) COMPUTED I.S. $= \frac{1}{3}\left[I.S.(Fe\text{-}Al) + I.S.(Fe\text{-}Ge) + I.S.(Fe\text{-}La)\right]$

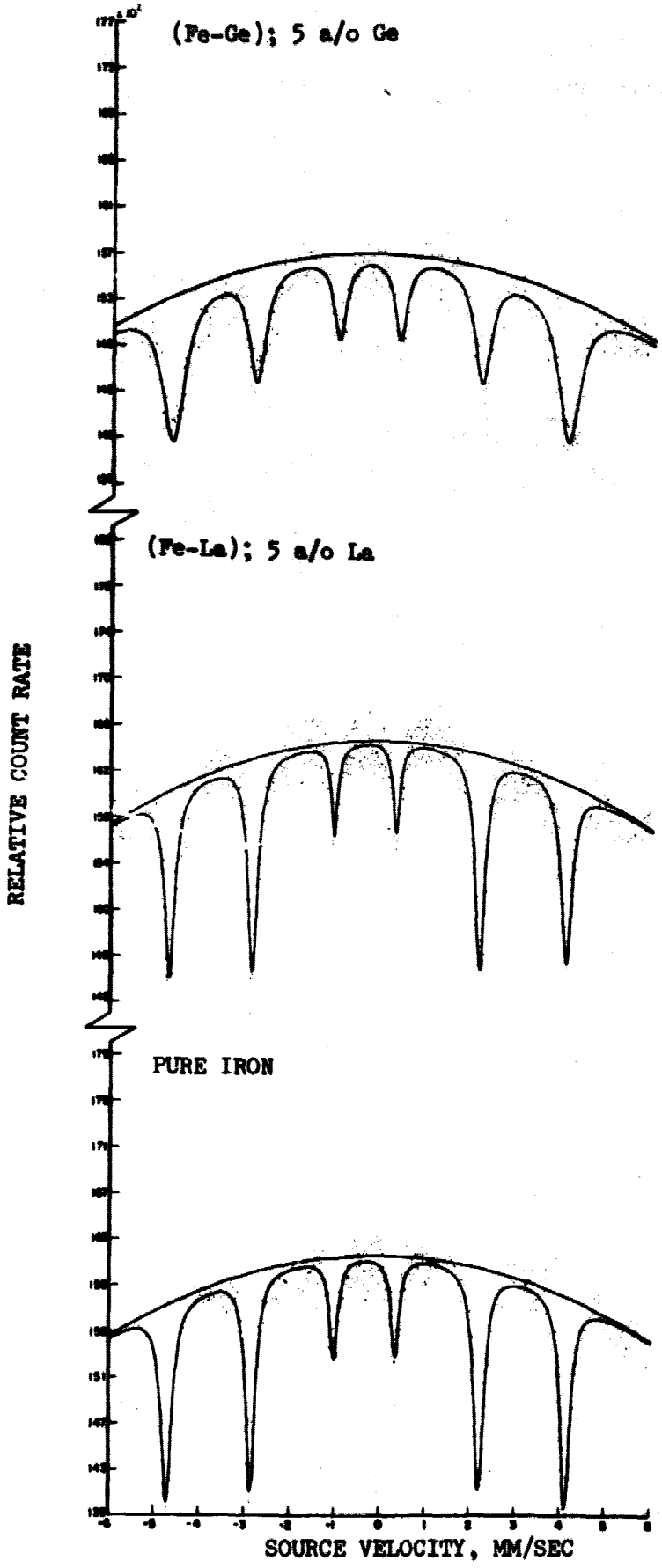

FIGURE 1.- THE EFFECTS OF LOW LEVEL CONCENTRATION (5%) OF La AND Ge IN Fe ON ITS MOSSBAUER SPECTRUM.

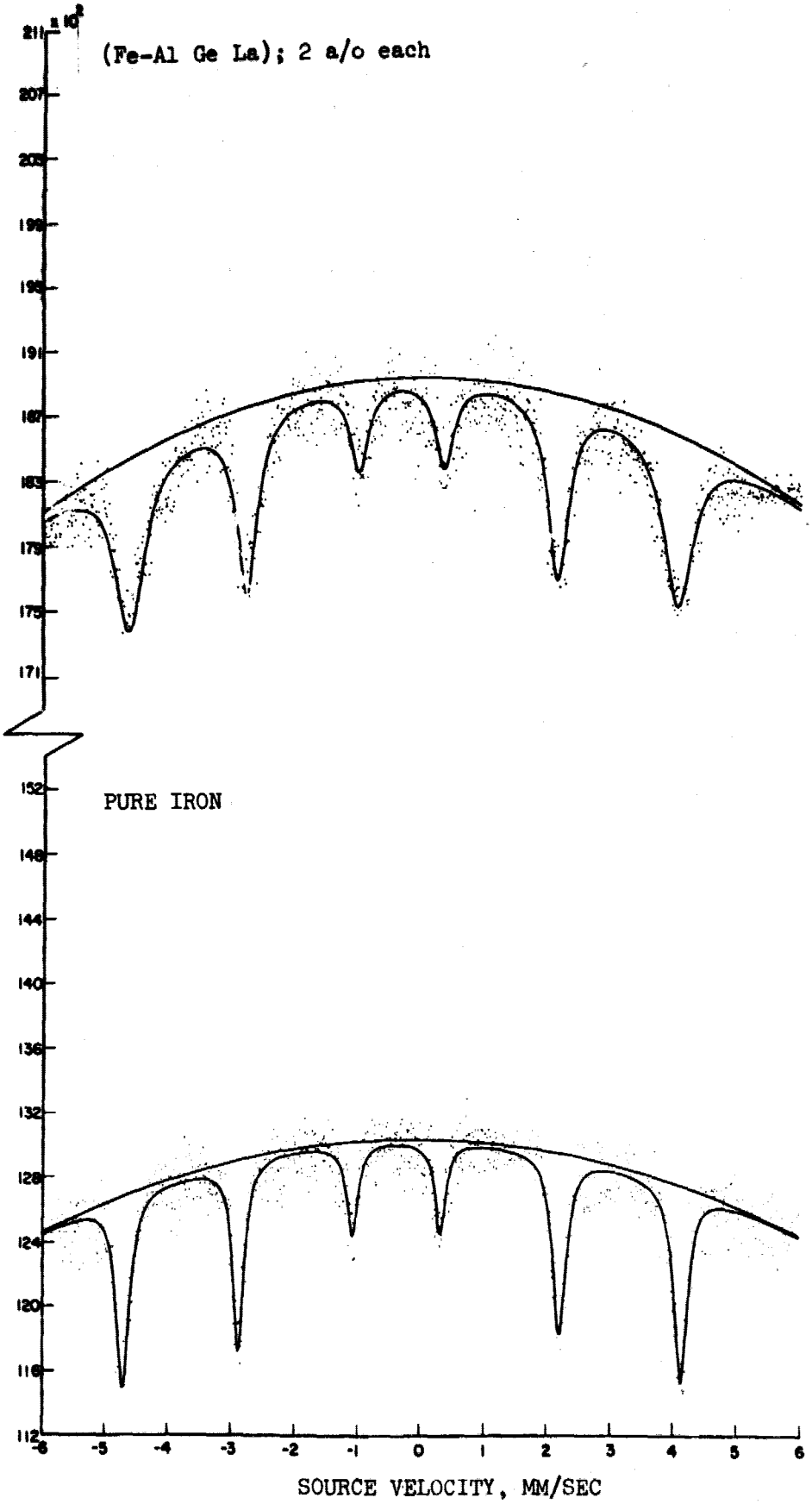

FIGURE 2.- THE EFFECTS OF SIMULTANEOUS LOW LEVEL CONCENTRATIONS (2% EACH) OF Al, Ge, AND La IN Fe ON ITS MOSSBAUER SPECTRUM.

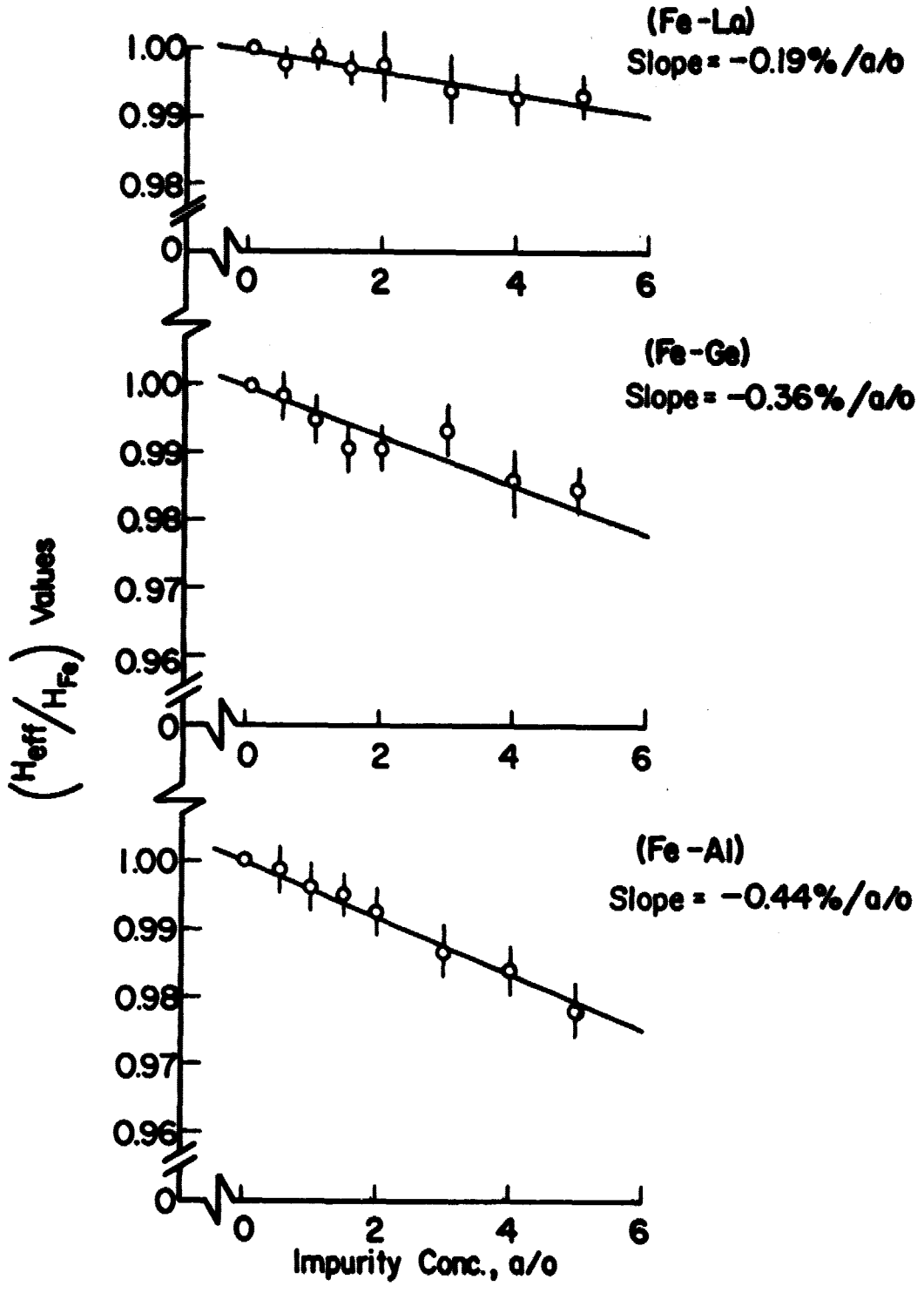

FIGURE 3.- VARIATION OF HYPERFINE FIELD AS A FUNCTION OF IMPURITY CONCENTRATION IN DILUTE BINARY ALLOYS OF IRON.

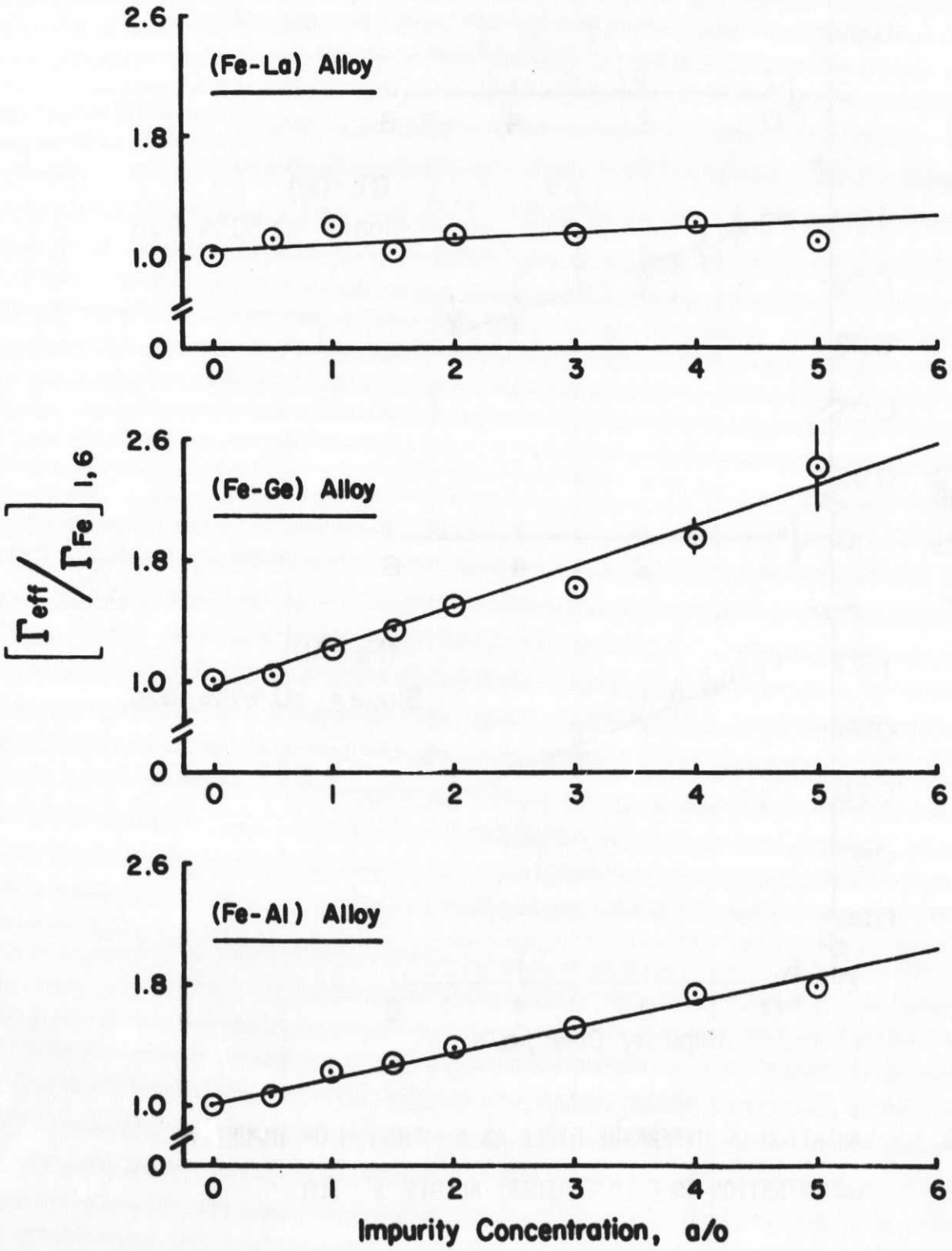

Figure 4. Variation of outer peak widths as a function of impurity Concentration in Dilute Binary Iron Alloys.

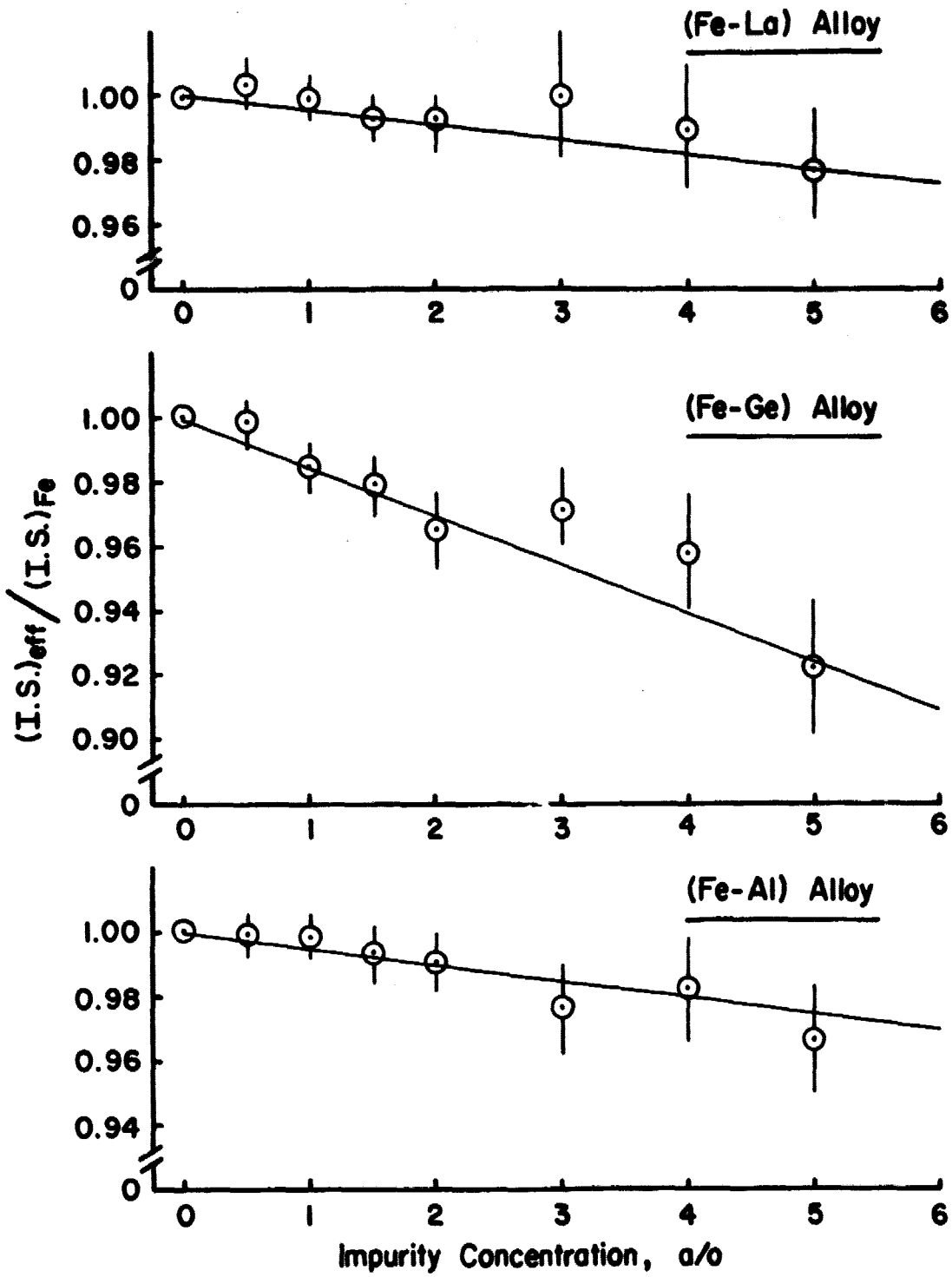

Figure 5. Variation of Isomer Shift as a function of Impurity Concentration in Dilute Binary Iron Alloys.

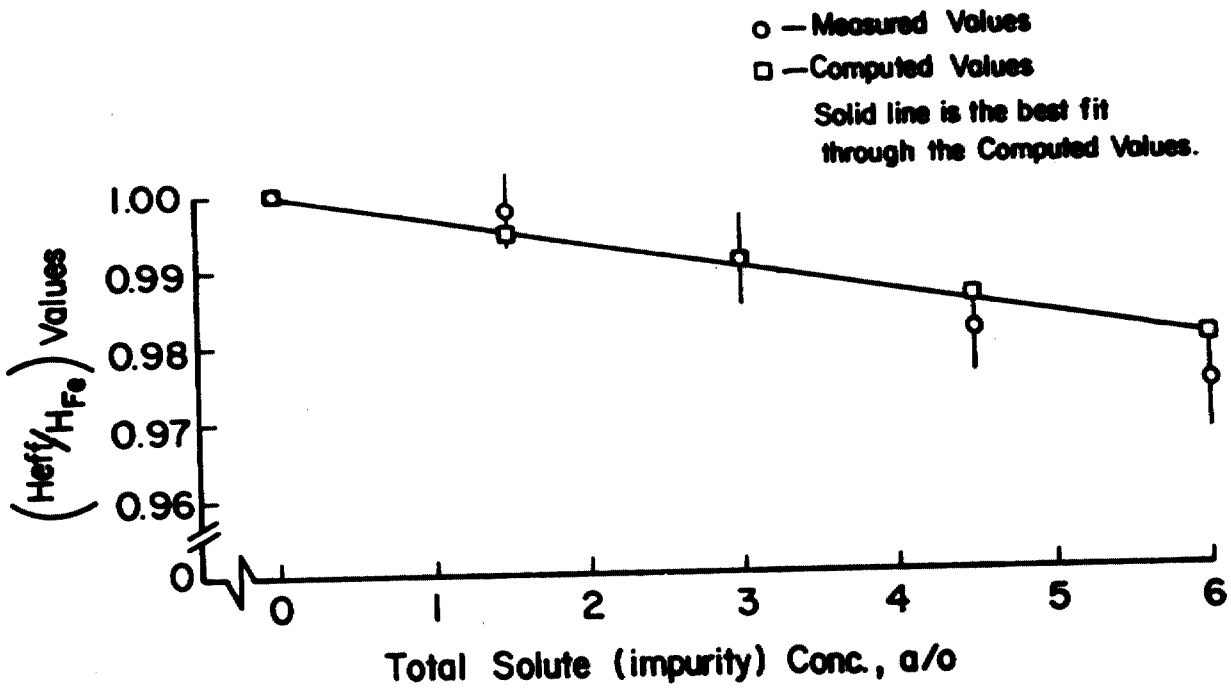

FIGURE 6.- COMPARISON BETWEEN THE COMPUTED AND THE MEASURED VALUES OF (H_{EFF}/H_{FE}) FOR DILUTE (Fe-AlGeLa) ALLOYS.

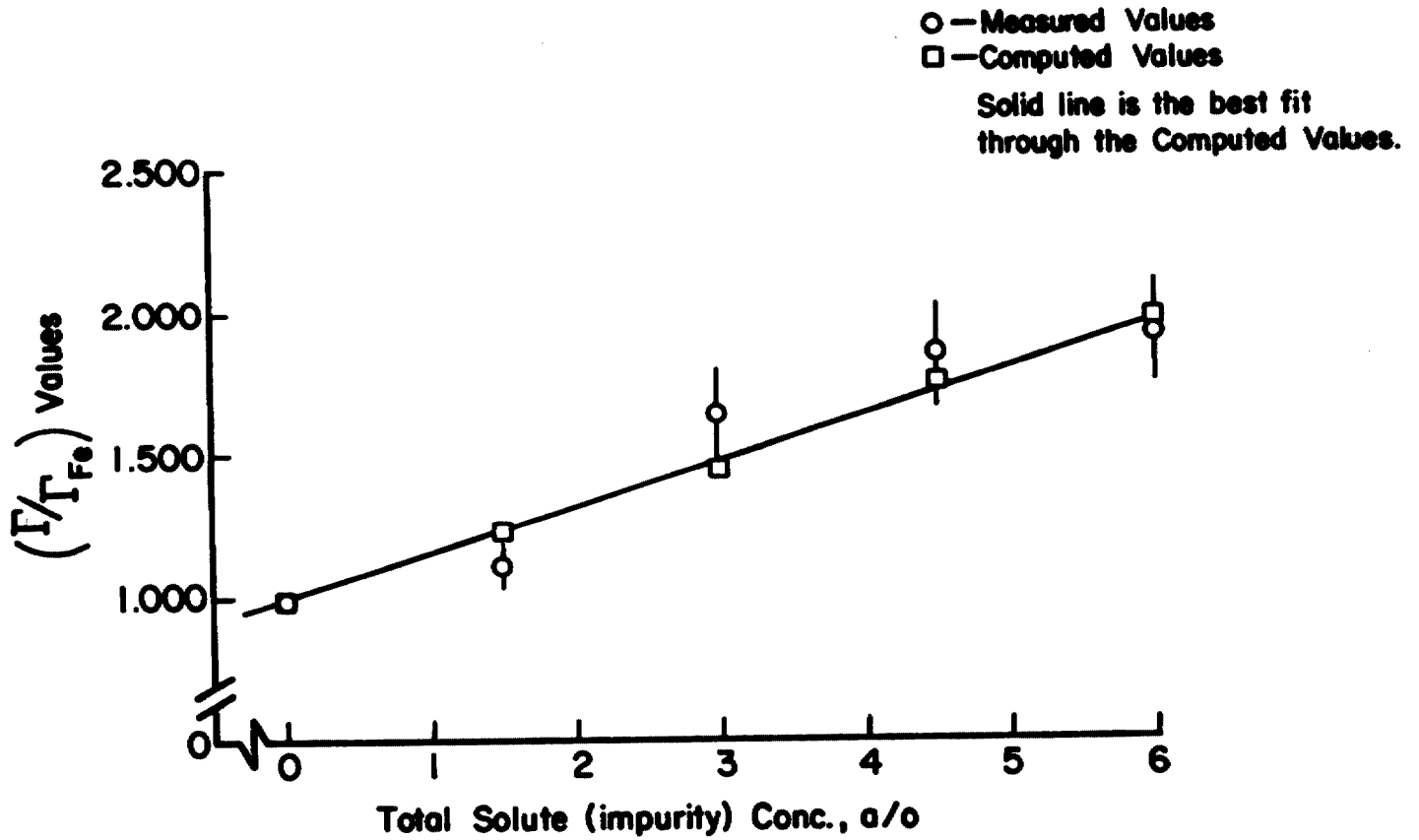

FIGURE 7. - COMPARISON BETWEEN THE COMPUTED AND THE MEASURED VALUES OF $\left(\Gamma/\Gamma_{Fe}\right)$ FOR DILUTE (Fe-AlGeLa) ALLOYS.

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15. Supplementary Notes

Data presented in this report indicate that the electronic and magnetic perturbations of simultaneously present dilute substitutional impurities in iron are additive.

16. Abstract

We have recently completed a study of Mossbauer parameters of dilute quaternary (Fe-XYZ) alloys where X, Y, Z, are Al, Ge, and La, respectively. A comparison between the computed and the observed values indicates that changes in effective hyperfine field (-0.37 $\pm$ 0.04% per a/o), outer peak widths (+16.30 $\pm$ 0.65% per a/o), and the isomer shift (-0.90 $\pm$ 0.04% per a/o) predicted on the basis of binary alloys of the individual impurities are equal - within experimental errors - to those obtained in quaternary (Fe-XYZ) alloys. This result indicates that the effects of simultaneously present dilute impurities are additive, independent of their respective electronic configurations.

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