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Gamma-ray Compton Profiles for Fe-Ti and Fe-Ti-H Amorphous Alloys*

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Synopsis

Compton profile measurements using 59.54 γ -ray source have been performed for Fe-Ti and Fe-Ti-H amorphous alloys. It is found that the electronic structure of Fe-Ti amorphous alloy is blurred by the vitrification, compared with the crystalline one, and that hydrogen atoms in Fe-Ti amorphous alloy prefer to donate valence electrons to the conduction band of the host lattice.

I. Introduction

Compton scattering technique of high energy γ -ray photons has been well established as a useful tool to investigate the electronic structure of matter¹⁾. This technique is not limited in principle from the state of the condensed form. In fact, this technique has been successfully applied to liquid metals²⁾ and amorphous alloys³⁾.

One of features of Compton profile measurement lies upon the fact that the Compton profile $J(Q)$ reflects the ground state wave functions of valence electrons which play important role in the bonding mechanism. This technique is particularly useful to investigate hydrogen state in the hydride, because the electrons from hydrogen atoms mainly participate in the form of bonding and anti-bonding state, contributing to the low momentum region of Compton profile and

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therefore the momentum distribution of electrons from hydrogen atoms is reflected in low momentum region of Compton profile.

Fe-Ti crystal is one of most promising materials for hydrogen storage⁴⁾. Recently it has been known that binary $\text{Fe}_{100-x}\text{Ti}_x$ alloys can be vitrified in the wide composition range of $20 < x < 75$ by vapour quenching method⁵⁾ and Fe-Ti amorphous alloys exhibit good hydrogen storage properties⁶⁾. In order to understand the solubility of hydrogen in Fe-Ti amorphous alloys, we measured Compton profiles for amorphous alloys of $\text{Fe}_{61}\text{Ti}_{39}$, $\text{Fe}_{54}\text{Ti}_{46}$ and $\text{Fe}_{61}\text{Ti}_{39}\text{H}_{16}$.

II. Experimental

Fe-Ti amorphous alloys were deposited on a water-cooled copper substrate into thick plate with the thickness of about $400 \mu\text{m}$ using a DC sputtering apparatus⁷⁾. The chemical composition of the sputtered film was determined for $\text{Fe}_{61}\text{Ti}_{39}$ and $\text{Fe}_{54}\text{Ti}_{46}$ amorphous alloys by means of electron probe micro analysis. For comparison, $\text{Fe}_{53}\text{Ti}_{47}$ crystalline alloy was also prepared. Hydrogen atoms were loaded into $\text{Fe}_{61}\text{Ti}_{39}$ amorphous alloy at temperature of 100°C and the hydrogen pressure of 35 kg/cm^2 . The content of hydrogen was determined to be $\text{Fe}_{61}\text{Ti}_{39}\text{H}_{16}$ by both the measurements of hydrogen gas pressure and sample weight.

The experimental set-up used in this study was essentially similar to that described in ref.[2]. 59.54 keV photons from 1 curie ^{241}Am γ -ray source were impinged on the sample and the inelastically scattered γ -rays were detected by a pure Ge solid state detector through the scattering angle of 165° . The energy resolution of the detector was 336 eV at the incident energy. The Compton profile was obtained in terms of the generalized least squares method⁸⁾ after various corrections. According to the deconvolution theory of generalized least squares method, theoretical Compton profile $J_{\text{th}}(x)$ can be compared after convoluting with the residual instrument function $g(x)$ as follows,

$$J^*_{\text{th}} = \int J_{\text{th}}(x')g(x-x')dx'. \quad (1)$$

The present $g(x)$ is given in Fig.1 and Table I. The data processing procedure is fully described in ref.[2].

Table I. The residual instrument function(RIF) against channel number, corresponding to 21.23 eV/channel in energy or 0.0334 a.u./channel in momentum. The parameters of the generalized least squares method used in this study are $k=3$, $\lambda=300$.

channel	RIF	channel	RIF	channel	RIF	channel	RIF
0	0.07618	1	0.07534	2	0.07304	3	0.06932
4	0.06431	5	0.05821	6	0.05127	7	0.04373
8	0.03587	9	0.02796	10	0.02027	11	0.01304
12	0.00648	13	0.00074	14	-0.00405	15	-0.00782
16	-0.01055	17	-0.01227	18	-0.01303	19	-0.01296
20	-0.01216	21	-0.01078	22	-0.00899	23	-0.00693
24	-0.00475	25	-0.00259	26	-0.00057	27	0.00121
28	0.00269	29	0.00384	30	0.00461	31	0.00503
32	0.00510	33	0.00487	34	0.00440	35	0.00373
36	0.00294	37	0.00207	38	0.00120	39	0.00037
40	-0.00038	41	-0.00101	42	-0.00150	43	-0.00185
44	-0.00205	45	-0.00211	46	-0.00204	47	-0.00186
48	-0.00160	49	-0.00128	50	-0.00092	51	-0.00056
52	-0.00021	53	0.00011	54	0.00038	55	0.00059
56	0.00075	57	0.00084	58	0.00087	59	0.00086
60	0.00079	61	0.00068	62	0.00055	63	0.00041
64	0.00026	65	0.00011	66	-0.00002	67	-0.00014
68	-0.00023	69	-0.00030	70	-0.00034	71	-0.00036

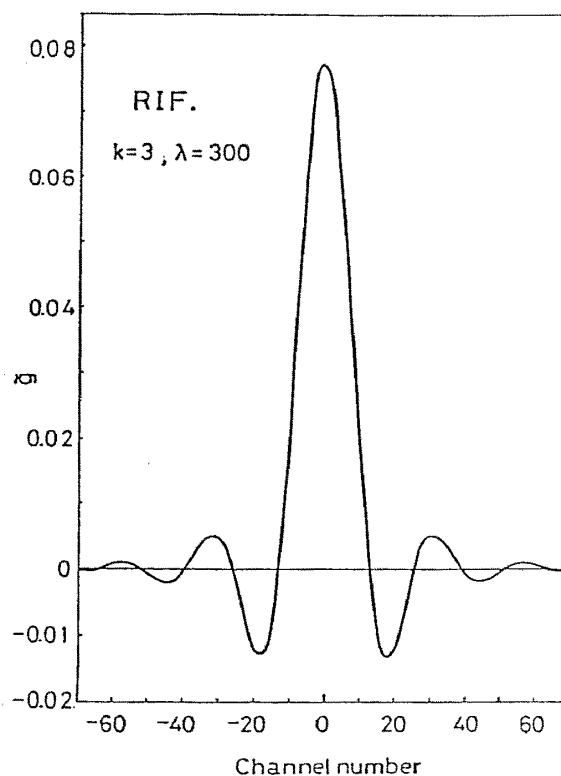


Fig.1. The residual instrument function $g(x)$ as a function of channel number. The numerical figures are given in Table I.

III. Results and Discussion

Figure 2 shows experimental $J(Q)$ for Fe-Ti amorphous and crystalline alloys. Each profile is normalized to give the same area with the theoretical profile by Biggs et al⁹⁾ in the range from $Q = -6.0$ a.u. to $Q = 6.0$ a.u. after the multiple scattering correction. It can be seen that the peak height $J(0)$ for $\text{Fe}_{54}\text{Ti}_{46}$ amorphous alloy gives a slightly higher value than that for $\text{Fe}_{53}\text{Ti}_{47}$ crystalline alloy.

In order to make the situation more clear, difference profile $\Delta J(Q)$ between the alloy itself and the weighted sum of its constituents is defined as follows,

$$\Delta J(Q) = J_{\text{alloy}}(Q) - (c_{\text{Fe}}J_{\text{Fe}}(Q) + c_{\text{Ti}}J_{\text{Ti}}(Q)) \quad (2)$$

where c denotes atomic concentration of the constituent in the alloy.

Figure 3 shows $\Delta J(Q)$ thus obtained, using Compton profiles for pure Fe and pure Ti from Yoda¹⁰⁾. All the curves display similar behaviour against the momentum Q ; $\Delta J(Q)$ shows negative values around $Q = 0$ and turn to positive around $Q = 1.0$ a.u., approaching zero after then. The overall similarity of $\Delta J(Q)$ behaviour between amorphous and crystalline alloys, though $\Delta J(0)$ values are slightly different, suggests that the bonding nature in the amorphous alloy is essentially similar to that in the crystalline counterpart. Compton profile of FeTi polycrystalline alloy was already reported by Lasser et al¹¹⁾ and the best agreement between theory and experiment was obtained for a configuration $\text{Fe } 3d^6 4s^2 - \text{Ti } 3d^2 4s^2$ in terms of the renormalized-free-atom model. Band structure calculations^{12,13)} have shown that there is a charge transfer of valence electrons from Ti to Fe. These calculations were performed in the energy space, so that the Compton profile results could not answer this prediction. Even at the present stage, it is still not clear whether the present $J(Q)$ and $\Delta J(Q)$ in Figs. 2 and 3 support the predicted tendency. Calculations of electron wavefunctions in r -space or Q -space is required.

Figure 4 shows a comparison of $J(Q)$ between $\text{Fe}_{61}\text{Ti}_{39}$ and $\text{Fe}_{61}\text{Ti}_{39}\text{H}_{16}$ amorphous alloy. Difference profiles $\Delta J_{\text{H}}(Q)$ defined as follows,

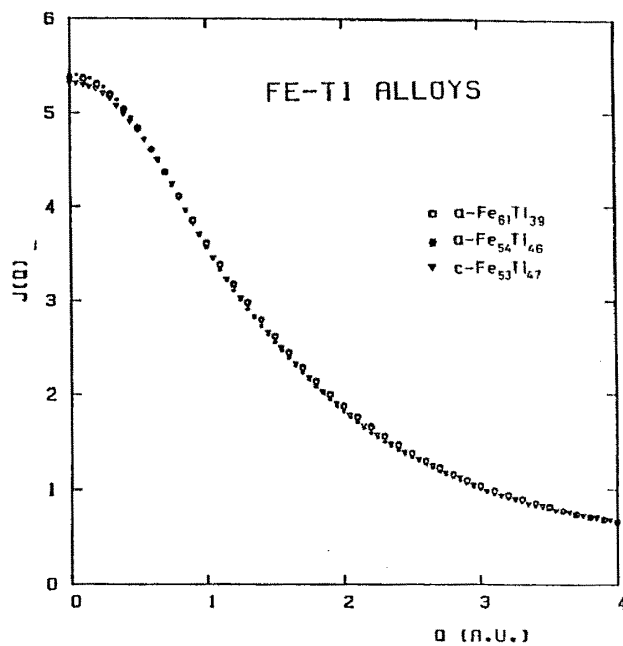


Fig.2. Compton profiles for $\text{Fe}_{61}\text{Ti}_{39}$ amorphous, $\text{Fe}_{54}\text{Ti}_{46}$ amorphous and $\text{Fe}_{53}\text{Ti}_{47}$ crystalline alloy, the area of which are normalized to give the theoretical values⁹⁾ of 21.09, 20.87 and 20.85, respectively, in the range from $Q = -6.0$ to $+6.0$ a.u.

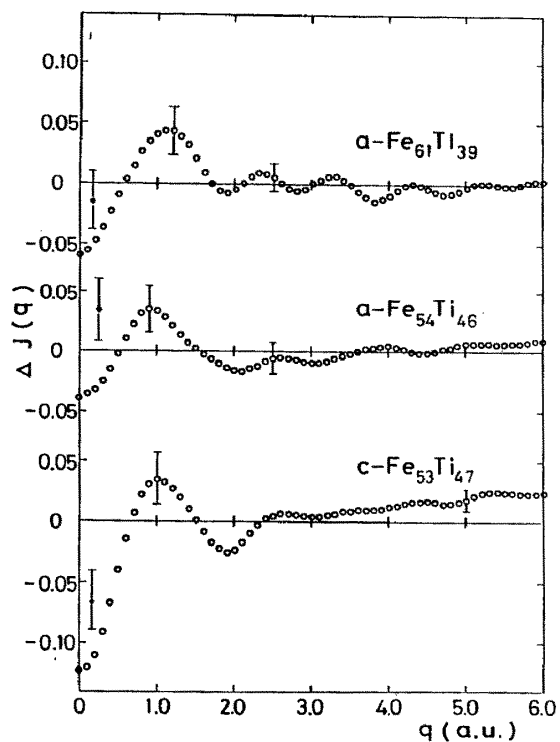


Fig.3. Difference Compton profiles $\Delta J(Q)$ for $\text{Fe}_{61}\text{Ti}_{39}$, $\text{Fe}_{54}\text{Ti}_{46}$ amorphous alloys and $\text{Fe}_{53}\text{Ti}_{47}$ crystalline alloy(see text).

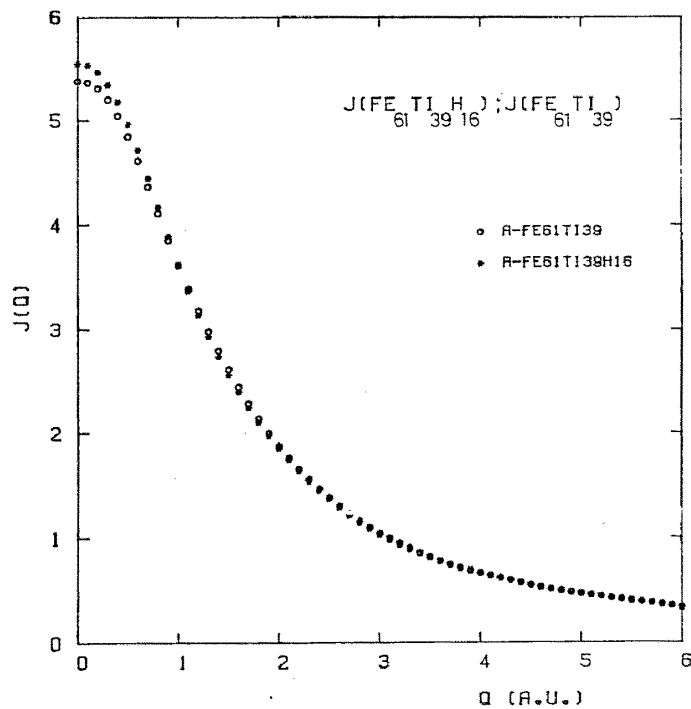


Fig.4. Compton profiles for $\text{Fe}_{61}\text{Ti}_{39}$ and $\text{Fe}_{61}\text{Ti}_{39}\text{H}_{16}$ amorphous alloys.

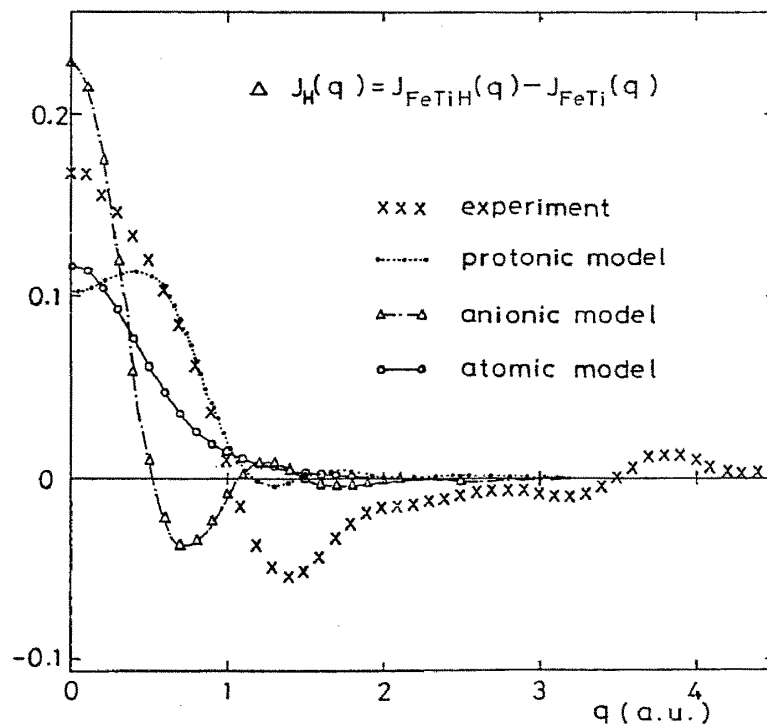


Fig.5. Comparison of $\Delta J_H(Q)$ between experimental profile and some model profiles(see text).

$$\Delta J_H(Q) = J_{\text{hydride}}(Q) - J_{\text{alloy}}(Q), \quad (3)$$

are compared between the experimental profile and some model profiles in Fig.5. From Figs.4 and 5, the effect of hydrogen can be seen to appear at small momenta less than $Q = 2$ a.u..

In order to understand the hydrogen state, three models are compared in Fig.5. One is the protonic model which was already used for V-H and Pd-H alloys¹⁴⁾. In this model, it was assumed that the hydrogen donates its electron to the conduction band of the host lattice and that the Compton profile of the valence electrons is increased proportional to the number of conduction electrons. In this paper, we take free electron model as the valence electron. One of the "protonic" model is shown in Fig.5, when taking 1.5, 1.3 and 1.0 for Fe, Ti and H, respectively, as the number of valence electrons per atom. The second model is an anionic model¹⁴⁾ which is based on the assumption that the hydrogen removes an electron from the conduction band of the host lattice and forms a negative ion H^- . The difference profile for this model is shown in Fig.5. The third model consists of an atomic model. In this model, the hydrogen atom exists, being isolated from the host lattice. The difference profile for this model is given as $\Delta J^0(Q) = 0.16 * J_H(Q)$, where $J_H(Q)$ shows the free atom profile given in ref.[9]. All the model profiles are convoluted by the residual instrument function $g(Q)$ given in Table I. It is found that the "protonic" model gives rather good agreement among the three models. However, the "protonic" model can not explain the experimental $\Delta J(Q)$ around $Q = 0$ and 1.3 a.u. . In order to quantitatively understand the hydrogen state in the amorphous alloy, it is necessary to have more sophisticated model which takes into account the solid state effects, such as bonding and anti-bonding hybrids. Further study on this line is now under way.

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