

CHAPTER VI

DISCUSSION

6.1 Mössbauer Spectrometer.

The use of a MCA in the Mössbauer constant acceleration spectrometer permits the measurement of a complete spectrum without readjustment, as such is well adapted to long experiments. In this spectrometer the reference signal was derived from the time sweep of the MCA, meaning the relation between the channel number and velocity is linear. Two precautions in this design should be considered.

(1) The triangular waveform is only available as long as the analyzer is in the time sweep. Stopping the analyzer without switching off the feedback gain (S_1 in Fig. 3.3) beforehand can result in damage to the drive coil of the transducer.

(2) The triangle amplitude will change when varying the time sweep period. These problems do not exist with the free running triangular wave generator described by Cohen (1966). Using a free-running oscillator as reference generator requires external synchronization of the time sweep of the MCA such as described by Kainal, et al. (1968).

It is important to connect the 0.1 μ F capacitor parallel to the potentiometer at the velocity-range position of Fig. 3.3 in order to attenuate the high frequency content in the input velocity reference voltage. This reduces the problem of high frequency oscillation which was encountered in controlling of the feedback system. It is then better to use the reference signal having lower high-frequency content.

In the operation, the following problems can be observed.

(1) If the transducer oscillates violently at a low frequency, the feedback system has the wrong polarity and the loud-speaker leads should be reversed.

(2) If the system has an oscillation at several KHz at all settings of the feedback gain, the bandwidth and the feedback gain should be reduced at the same time. The optimum setting depends somewhat on the high frequency content of the reference signal. For a sine wave the bandwidth can be reduced and the gain increased.

To restrict the amplitude of the reference signal, high frequencies are necessary so that the higher velocities can be obtained. The maximum open loop transfer curve of the transducer lies at the fundamental resonance of the transducer. Clearly, the fundamental frequency of the reference signal should be close to this resonance to give the best regulation of the motion. The resonance frequency of the transducer depends mainly on the mass of the moving part and the hardness of the guide springs. It can be increased by using the harder guide springs, or using a pair of springs on each side, separated by a thin space. For high velocities, a sinusoidal signal is the recommended reference waveform. Sinusoidal drive motion has the advantage of keeping inertial forces to a minimum since no sudden changes in acceleration occur and less power has to be delivered by the power amplifier. A small disadvantage is that, since the analyzer time sweep is linear, the

relation between channel number and velocity is non-linear. 1

In the spectrometer, each velocity is reached twice during one full sweep and thus the velocity spectrum is actually stored twice in the MCA (see Fig. 5.5 and 5.6). The spectrum in the second half is the mirror image of the first half. To obtain the true velocity spectrum, the counts in the channel 0, 1,, 511 have to be added to those in the channel 1023, 1022, ..., 512. Since the velocity pick-up signal was not actually in phase with the reference signal, it lags about 160 μ s which can be seen by the oscilloscope. Then the data in the channel 515 corresponded to those of the channel 516 of the same velocity and so on, must be added. This "folding procedure" also eliminated geometrical effect on the base line (see Fig 5.5 and 5.7). This geometrical effect is caused by the fact that each velocity is coupled to a certain source position during the time intervals that each channel is opened. Thus a variation of the count rate with velocity will occur even in the absence of resonance. A poor alignment of source, absorber and detector can produce a highly asymmetric geometry effect that is eliminated in folding.

The linearity of the velocity depends on

- (1) the linearity of the triangular reference signal,
- (2) the degree to which the transducer can be forced to follow the reference signal, and
- (3) the linearity of the velocity sensor.



The linearity of the reference signal is determined by the accuracy of the integrating circuit which is approximately 10^{-4} (Kalvius, et al. 1972). A measurement of nonlinearity due to (2) was obtained from the ratio of the error voltage in the feedback loop to the reference voltage. This ratio was 0.15 % for the experimental reported here. The velocity sensor (pick-up coil and magnetic field) of the transducer contributed the major deviation from linearity of the spectrometer. The main contribution depends on the uniformity of the magnetic field within the pick-up coil. It can be seen from Fig. 5.7 that the velocity scale is practically linear over the entire range of the 512 channels, with a systematic deviation of ± 0.5 %.

To reduce the geometrical effect and the problems with pick-up coil linearity, higher frequencies are recommended for a reference signal because of the small amplitude motion. Its disadvantages are for one is the need for higher inertial forces, which require a more rigid source construction. Secondly, it requires shorter operating times of the channels in the time mode operation of the MCA. Assuming 1000 channels and about 50 Hz driving frequency, the dwell time in each channel would be $t_s = 20 \mu s$. In the time mode t_s cannot be made deliberately short because several logic procedures have to be carried out in switch form one channel to the next. After the channel advance pulses arrive, the analyzer closes its input for all arriving nuclear counts for the time span t_m . During t_m the number of counts stored during the last channel opening time are transferred from a register scaler into the appropriate location of the memory, the scaler is reset and the channel number advanced. After this the input is opened again to accept nuclear counts. This shows that the multiscaler

can be used for storage in each channel only during the time span $t_s - t_m$. The time t_m is called the memory cycle time and is fixed in the ND 2400 at about 2.25 μ s. Thus dead time losses will become serious if

$$t_s \lesssim 5 t_m.$$

6.2 Observed Spectrum

Figure 5.5 shows the Mössbauer spectrums of ^{57}Fe in a natural iron absorber of 0.025 mm thickness. It should be pointed out here that the asymmetric appearance of Fig. 5.5 (a) is due to the geometrical effect which has been mentioned in section 6.1. By folding the data in the manner described in section 4.2, the geometrical effect can be eliminated to give Fig. 5.7. The velocity calibration constant was determined to be 0.0407 ± 0.0002 mm/sec-channel by the method explained in Chapter V.

Figure 5.6 shows the Mössbauer spectrums of ^{57}Fe in a CoFe_2O_4 absorber in the form of large chunks and in a CoFe_2O_4 absorber in the form of fine powder. It should be noted that the absorption lines of ^{57}Fe in the large chunk absorber is broader than the same absorption lines in the fine powder absorber. This line broadening arises because of the thickness of the absorber and the greater nonuniformity in the density of CoFe_2O_4 when it is in the large chunk form. Even sharper absorption lines could probably have been obtained if the fine powder had been compressed.

6.3 Theoretical Discussion.

It has been pointed out by Tang (1977) that if one looks at the field acting on the B site Fe^{3+} ion,

$$\langle H_{eff}(Fe_A^{3+}) \rangle_r = \text{first term} - x N_A \left[J^{(4)} \langle \langle Fe_A^{3+} \rangle \rangle_r - J^{(2)} \langle \langle Co_A^{2+} \rangle \rangle_r \right] \quad (2.3.13b),$$

one sees that the above equation is consistent with the observation of Sawatzky, et al. (1969) if $J^{(4)} \langle \langle Fe_A^{3+} \rangle \rangle_r > J^{(2)} \langle \langle Co_A^{2+} \rangle \rangle_r$.

Sawatzky et al. observed that the field decreases as Co^{2+} ions replace Fe^{3+} ions on the A site. (This corresponds to an increase of x).

Looking at the field acting on the A site Fe^{3+} ion, eq. (2.3.13a) and again assuming that $J^{(4)} \langle \langle Fe_B^{3+} \rangle \rangle_r > J^{(3)} \langle \langle Co_B^{2+} \rangle \rangle_r$,

$$\langle H_{eff}(Fe_A^{3+}) \rangle_r = \text{first term} + \frac{x}{2} N_B \left[J^{(4)} \langle \langle Fe_B^{3+} \rangle \rangle_r - J^{(3)} \langle \langle Co_B^{2+} \rangle \rangle_r \right] \quad (2.3.13a),$$

one observed that a decrease of x would mean that the field on Fe_A^{3+} would be reduced. This is also in keeping with Sawatzky et al.'s observation that the field on A site decreases as Fe^{3+} ions on the B site are replaced by Co^{2+} ions. (An increase of Co^{2+} ions on the B site corresponds to a decrease of Co^{2+} ions going on to the A site, since the total number of Co^{2+} ion is constant).

It is also important to note that the theory has treated $J_{ij}^{(2)}(Co_i^{2+}, Fe_j^{3+}) \neq J_{ij}^{(3)}(Fe_i^{3+}, Co_j^{2+})$, unlike the molecular field study of Stephenson (1972) which considered them equal. The implication of this is that the same % change in the concentration of Co^{2+} on a particular

sublattice would not have to produce a similar change of the magnetic field on the other sublattice. This effect was pointed out by Evans (1968), who mentioned that a 4 % change in the concentration of Co^{2+} ion on the A site produced a change in the field on B site while a 50 % change in Co^{2+} ion on the B site did not produce a similar change on the A site.

6.4 Future Work.

Now that we have been able to construct an instrument for studying the local hyperfine fields within a ferrimagnetic spinel compound or any other type of magnetic compounds, it will now be possible to study the effects on the local fields produced by variation of any parameters of the crystal structure of the compound or by changing the types of ions which occupy different sites. By applying a magnetic field at the absorber, and improving of the preparation of the powder absorber for the Mössbauer experiment, we should be able to resolve the A - and B - site Fe^{3+} Mössbauer absorption peaks. In addition, the experiment could be performed at the liquid nitrogen temperature. Thus the resolution of the spectrometer could be increased further.