Mössbauer effect in the Co_2MnSn alloy under pressure

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The Mössbauer effect method was used in an investigation of the pressure dependences of the isomeric shift ϵ and effective magnetic field H at the Sn^{119} nuclei in the ferromagnetic alloy Co_2MnSn at room temperature. It was found that the changes in these quantities were $\Delta H/H = -3\%$ and $\Delta\epsilon = -0.02$ mm/sec at 14 kbar. The experimental results were analyzed on the basis of alternative models concerned with the mechanisms of the appearance of effective magnetic fields at the nuclei of nonmagnetic atoms in ferromagnetic matrices.

The effective magnetic fields at the nuclei of nonmagnetic atoms in ferromagnetic matrices have been the subject of intensive experimental and theoretical investigations. In particular, the Mössbauer effect studies have revealed that the magnetic fields at the tin nuclei are not proportional to the magnetic moments of the matrices and can have even different signs in different matrices.[1] There is as yet no consistent theory of these fields. Only a few attempts have been made to explain qualitatively the fields in question. They include two alternative models for the description of these fields. According to the first model,[2,3] magnetic fields are produced mainly because of the contact interaction between the nuclei of the impurity atoms and the polarized conduction electrons of the ferromagnetic matrix. According to the second model,[4,5] a dominant role is played by the internal shells of the nonmagnetic atoms, which are polarized by the surrounding ferromagnetic matrix atoms.

To decide which of these models is correct, we must carry out experiments in which the consequences of these models manifest themselves in different ways. Such experiments may include investigations of the influence of pressure on the effective magnetic fields of nonmagnetic atoms. For example, if the first model is correct, we should find that the pressure-induced change in the effective magnetic field is proportional to the density of the s conduction electrons at the nucleus of a nonmagnetic atom; in this case, it is necessary to allow for the change in the magnetic moment of the matrix under pressure. If the second model is correct, the increase in the field under pressure should be negative for any matrix. These points will be discussed later.

The present authors are aware only of the investigation of Möller,[6] who determined the influence of pressure on the effective magnetic field at the nuclei of tin impurities in iron. However, the large random errors made it impossible to establish a correlation between the changes in the field and the isomeric shift. We shall report the results of an investigation of the influence of pressure on the isomeric shift and the effective magnetic field at the Sn^{119} nuclei in the Co_2MnSn ferromagnetic alloy. This alloy was selected because of: 1) the high probability of the Mössbauer effect, which made it much easier to determine the parameters of the Mössbauer spectra at high pressures; 2) the structure and properties of the Co_2MnSn alloy were close to the Heusler alloys, known to be described satisfactorily by the Friedel theory of the spin density oscillations.

EXPERIMENT

Samples of the Co_2MnSn alloy were prepared by fusing together the calculated amounts of the components at 1400°C in an argon atmosphere and quenching by contact with copper. The Mössbauer effect was enhanced by using tin enriched to 91% with the Sn^{119} isotope. The ingots which ground into powders and then annealed at 700°C for 5 h. The powders were made up into absorbers $\sim 25 mg/cm^2$ thick. The spectra were determined using a constant-velocity Mössbauer spectrometer with SnO_2 source of ~ 15 μCi activity. A hydrostatic pressure was produced in a chamber similar to that described by Panyushkin.[7] The pressure-transmitting medium was a mixture of transformer oil and kerosene. The pressure was measured with two calibrated manganin resistance probes to within ± 3%. Each Mössbauer spectrum under pressure was recorded in two days. During this time, the pressure in the chamber fell by ~ 0.5 kbar. The absolute error in the determination of the pressure, indicated in Figs. 2 and 3 by the horizontal segments (this point is discussed later), included both errors.

RESULTS OF MEASUREMENTS AND DISCUSSION

Figure 1 shows the Mössbauer spectra of the Sn^{119} nuclei in the Co_2MnSn alloy under normal conditions (curve a) and under a pressure of 14 kbar (curve b). The random errors did not exceed the dimensions of the points in Fig. 1. The form and the parameters of the spectrum (curve a) were practically identical with those reported by Williams.[8] The average magnetic field at the Sn^{119} nuclei, expressed in kilooersteds, was deduced from the formula $\Gamma = 0.14H + 1.2$,[9] where Γ is the half-width of the spectrum (mm/sec). The spectra were analyzed by the least-squares method.

The pressure dependence of $-\Delta H/H$ is plotted in Fig. 2 in the form of $\Delta H = H(0) - H(p)$. This dependence was linear within the limits of the experimental error. The observed relative change in the field was fairly large. We should recall that, in the case of Fe^{57} in iron, this change was $\Delta H/H = 1\%$ at 100 kbar.[10] Figure 3 shows the pressure dependence of the change in the position of the center of gravity of the spectrum, $-\Delta\epsilon$. The relativistic shift under pressure could be ignored.[11] Therefore, Fig. 3 represents the reduction in the density of the 5s electrons at the Sn^{119} nuclei in the alloy. This effect could be explained in the same way as in the case of pure tin under pressure, i.e., it could be attributed to the screening of the spherically symmetric cloud of the 5s electrons of the tin atom by the 5p electrons.[11]

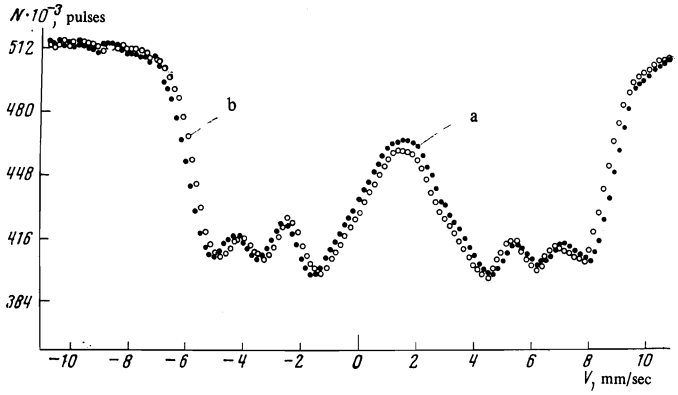

FIG. 1. Mössbauer spectra of the Sn^{119} nuclei in the Co_2MnSn alloy: a) P = 1 atm; b) P = 14 kbar.

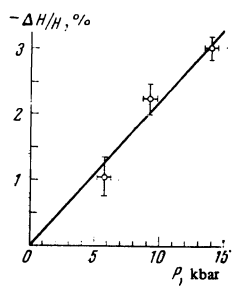

FIG. 2

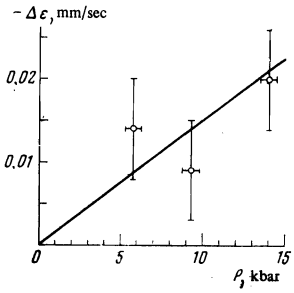

FIG. 3

FIG. 2. Relative change in the effective field at the Sn^{119} nuclei in the Co_2MnSn alloy under pressure.

FIG. 3. Change in the position of the center of gravity of the spectrum of the Sn^{119} nuclei in the Co_2MnSn alloy under pressure.

We shall now consider the change in the magnetic field on the basis of the two models mentioned above. According to the Caroli-Blandin model,[3] the effective field at the Sn^{119} nuclei is

$$H=\frac{8\pi}{3}\mu_B[|\psi_{\uparrow 5s}(0)|^2-|\psi_{\downarrow 5s}(0)|^2]np. \qquad (1)$$

Here, $|\psi_{\uparrow 5S}(0)|^2$ and $|\psi_{\downarrow 5S}(0)|^2$ are the probability densities of the 5s conduction electrons with oppositely directed spins; n is the number of the 5s electrons per one tin atom; p is the polarization of the 5s electrons.

It should be pointed out that Eq. (1) in the Caroli-Blandin paper[3] was used in quantitative calculations of H at the nuclei in some Heusler alloys employing the spatial distribution of the spin density of the conduction electrons. It was recently shown[12] that a similar calculation could not, at least in the case of the Cu_2MnSn and Ni_2MnSn alloys, give the positive magnetic field found experimentally without a detailed knowledge of the energy band structure and of the wave functions of the conduction electrons. In the case of the Co_2MnSn alloy, the field H at the Sn^{119} nuclei was positive and, moreover, there was a small localized magnetic moment at the Co atoms which was ignored in the calculations based on Eq. (1). For these reasons, we analyzed the consequences of Eq. (1) only qualitatively, ignoring the specific difficulties of the theory. In other words, we assumed–in accordance with the Caroli-Blandin theory–that the field H was entirely due to the contact interaction between the nucleus and the polarized 5s electrons of the conduction band.

According to Eq. (1), the change in H under pressure may be due to a change in the number of 5s electrons or a change in their polarization and density at the nucleus. At relatively low pressures, the change in the number n of these electrons can be ignored. It is also natural to assume that the polarization is proportional to the magnetic moment of the sample. The magnetic moment at a given temperature changes under pressure because of the influence of pressure on the Curie temperature T_C and on the magnetization σ_0 at T = 0°K. This is shown schematically in Fig. 4. In the case of the Co_2MnSn alloy, the influence of pressure on T_C can be ignored because $T_C \approx 800$°K and, the measurement temperature correspond effectively to the plateau of the Brillouin curve, i.e., $\sigma \approx \sigma_0$. There is no published information on the influence of pressure on σ_0. However, in the case of one of the Heusler alloys (Cu_2MnIn), it is known that $\Delta\sigma/\sigma_0\Delta P = -2.1 \times 10^{-3}$ kbar^{-1}.[13] We shall use this value as a rough estimate for our alloy.

We shall allow for the change in the density of the 5s electrons in the following way. We shall assume that, in the case of α-Sn, the electron configuration of the Sn atom is $5s^1 5p^3$[14] and the addition of one 5s electron increases the isomeric shift by 2.6 mm/sec. We thus find that the Sn configuration in the alloy is $5s^{0.85}5p^x$. Nothing definite can be said about the density x but this is not important in our discussion. According to the adopted model, 0.85 5s electrons produce a field H = +97.6 kOe at an Sn^{119} nucleus. Moreover, up to 50 kbar, the change in the electron density is proportional to the applied pressure.[11,15,16] Therefore, the reduction in the isomeric shift by 0.02 mm/sec at 14 kbar (Fig. 3) corresponds to $\Delta H/H = -1.5\%$, which represents about half the value found experimentally; the rest of the change $\Delta H/H$ can be attributed to the reduction in σ.

We shall now consider our results from the point of view of the Balabanov-Delyagin model,[4] according to which the field at the Sn^{119} nuclei is

$$H=H^+(r)-H^-(r), \qquad (2)$$

where $H^+(r)$ and $H^-(r)$ are large (in the absolute sense) contributions to the resultant field, and r is the distance from the Sn nucleus. It is important to point out that the function $H^-(r)$ varies more rapidly than $H^+(r)$. Therefore, when r is reduced by the application of a pressure, ΔH should be negative for any matrix, irrespective of the sign of H under normal conditions. The sign of ΔH thus agrees with that found experimentally. Unfortunately, the functions $H^-(r)$ and $H^+(r)$ are not known and there is no way of comparing the results quantitatively with this model. It should also be noted that ΔH, predicted by Möller,[6] is also negative.

The following conclusions can be drawn from our discussion. Although the results of our experiments are not in conflict with either of the two models, we are inclined to adopt the Balabanov-Delyagin explanation. The final answer on the correctness of one or the other model can be made by continuing similar investigations of H at the Sn^{119} nuclei in matrices for which |H| is

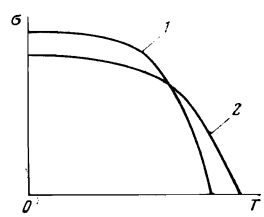

FIG. 4. Schematic temperature dependence of the magnetization: 1) P = 1 atm; 2) P > 1 atm. We can see that the magnitude and sign of the difference $\Delta\sigma = \sigma_2 - \sigma_1$ may vary with temperature.

small (for example, Co and Ni). If ΔH in these matrices is of the same order as in Co_2MnSn and Fe, the Caroli-Blandin model[3] should be rejected because $\Delta\sigma/\sigma_0\Delta P$ in the case of Co and Ni is considerably smaller than for the Heusler alloys.

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