INTERNAL MAGNETIC FIELDS AT Sn^{119} *NUCLEI IN HEUSLER ALLOYS*

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The Mössbauer effect was used to determine the magnetic field strengths at the Sn^{119} nuclei in the ordered ferromagnetic Heusler alloys Cu_2MnSn and $Cu_2Mn_{1.1}Sn_{0.9}$, and the values 200 ± 35 and 235 ± 35 kOe were obtained at liquid nitrogen temperature. Such relatively high values of the fields at a Sn^{119} nucleus supported the assumption of a possible indirect exchange interaction between Mn atoms via a nonmagnetic tin atom. In such a case, the magnetic field at tin nuclei would be governed mainly by the contribution of the polarization of the ion core and the polarization of the conduction electrons.

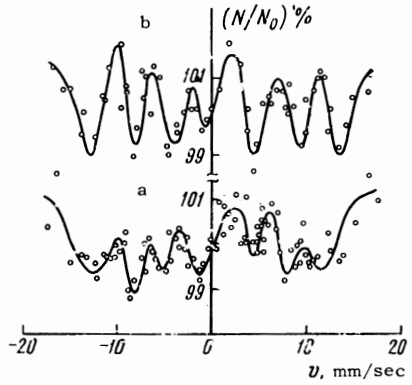
ORDERED ferromagnetic Heusler alloys belonging to the ternary systems Cu—Mn—Sn, Cu—Mn—Al, and Cu—Mn—In have been the subject of many investigations of the influence of the interatomic separation between the transition atoms of manganese on the nature of magnetic ordering. The crystal structure of ordered Heusler alloys can be described as an NaCl-type structure of Mn and Sn atoms with Cu atoms in the tetrahedral positions. Among the ferromagnetically ordered alloys of the systems Cu—Mn—Sn, Cu—Mn—Al, and Cu—Mn—In the highest value of the magnetization is observed in the compositions Cu_2MnSn, Cu_2MnAl, and Cu_2MnIn, in which the magnetic moment per atom of Mn is approximately $4\mu_B$, and in which it is usually assumed that nonmagnetic atoms of Cu, Sn, Al, and In do not carry magnetic moments.[1-3] However, Slater[4] has suggested that, in the aluminum Heusler alloy the copper atoms may make a contribution to the magnetization.

The nature of the interaction responsible for the ferromagnetism of these alloys is not yet known exactly. Since manganese atoms are separated by nonmagnetic atoms, it is assumed that the ferromagnetism of these alloys is evidently either due to an indirect exchange mechanism either via the conduction electrons[5] or due to a superexchange via nonmagnetic atoms.[2] The data from a neutron-diffraction investigation of the Cu_2MnAl alloys[2] fit better the mechanism of interaction via the conduction band, but they do not exclude the possibility of a superexchange. The mechanism of interaction via the conduction electrons[5] presumes the antiferromagnetic sign of the interaction between the 3d-shells of Mn, irrespective of the interatomic distance, and the ferromagnetic coupling between the conduction electrons and the 3d-shells. If the interatomic distances of the Mn atoms are sufficiently large, the dominant mechanism is the interaction via the conduction electrons; therefore, the spins of manganese atoms assume parallel alignment.

If we assume the mechanism of an indirect exchange interaction via the conduction electrons, the magnetic field at the tin nuclei in the Heusler alloy Cu_2MnSn should be due mainly to the contribution of the polarization of the conduction electrons because the contact and orbital contributions of nonmagnetic tin atoms should be zero and the contribution of the Lorentz field should be small and easily allowed for. Therefore, the measurement of magnetic fields at tin nuclei in Heusler alloys is of interest both in relation to the determination of the contribution of the polarization of conduction electrons to the magnetic field, and in gaining a better understanding of the ordering mechanism.

We determined the internal magnetic fields at the nuclei of Sn^{119} in the ordered ferromagnetic Heusler alloys Cu_2MnSn and $Cu_2Mn_{1.1}Sn_{0.9}$. A foil of metallic radioactive tin Sn^{119}* was used as the source of the resonance γ-radiation. The source and the absorber were at liquid nitrogen temperature. The alloys were prepared from electrolytic manganese, 99.99% pure copper, and 99.999% pure tin using the method described in[1].

After the samples had been quenched, annealed, pulverized and then heat treated, they were subjected to an x-ray diffraction analysis. The composition of the alloys was checked by chemical analysis. The values of the saturation magnetic moments of these compositions, measured by means of a pen-



Absorption spectra of γ quanta of 23.8 keV energy in the Heusler alloys Cu_2MnSn (a) and $Cu_2Mn_{1.1}Sn_{0.9}$ (b). (N/N_0)% is the relative absorption.

dulum balance, were practically identical with the values reported in[1]. The absorbers were prepared by the precipitation of a powder of an alloy onto an aluminum foil.

The absorption spectra were recorded using apparatus with a mechanical drive. Each of the absorption spectra (cf. figure) represented the superposition of two spectra, one of which consisted of six outer lines, and the other a central absorption peak. This interpretation was partly confirmed by the coincidence, within the limits of experimental error, of the values of the internal magnetic field calculated, from the six-line spectrum, for the distribution of the ground and excited states of the Sn^{119} nucleus. The broadening of the spectra was evidently due to the incomplete ordering of the samples (because of which there was a range of magnetic fields at the nuclei), possible quadrupole interaction, and mechanical vibrations at high velocities. The central peak was probably due to tin atoms located outside the center of the cube consisting of Cu atoms (because of the incomplete ordering of the alloy) or due to the presence of a second phase.

The magnetic fields at the tin nuclei were 200 ± 35 kOe for Cu_2MnSn and 235 ± 35 kOe for $Cu_2Mn_{1.1}Sn_{0.9}$. If one assumes the mechanism of magnetic ordering based on the interaction via polarized conduction electrons, these values are far too large. In fact, the magnetic field at Sn^{119} nuclei in a dilute (1%) solid solution of tin in iron was found to be 88 kOe.[6] This field was assumed to be due to the contribution of the polarization of conduction electrons. The contribution of the Lorentz field was, according to a calculation, equal to +7.5 kOe, but the contribution of the polarization of the ion core was not allowed for by Boyle, Bunbury, and Edwards.[6] In this case, the exchange field of magnetic electrons of neighboring transitions atoms polarizes a nonmagnetic atom, giving rise to unpaired s-electrons and a negative contribution to the magnetic field at the nucleus of the nonmagnetic atom. In any case, for an impurity atom of tin in iron this contribution should be greater than that for an atom of tin in Heusler alloys because the magnetic moments of neighboring iron atoms are larger than the magnetic moments of the manganese and copper atoms that are neighbors of tin even if it is assumed, in accordance with[4], that the 3d-shell of copper in Heusler alloys does not have the $3d^{10}$ configuration. Moreover, the distance from a tin atom to the nearest transition atom in a Heusler alloy is greater in a dilute solid solution of tin in iron. Therefore, the value of the magnetic field at tin nuclei in iron, which is about 88 kOe, can be taken as an approximate upper limit of the contribution of the polarization of conduction electrons to the magnetic field expected at a tin nucleus in a Heusler alloy.

The value of $\approx$200 kOe obtained by us indicates the presence of other contributions, the chief of which is probably the contribution of the ion core polarization.[7] Therefore, we can use the mechanism of an indirect superexchange between transition atoms of the alloy via a nonmagnetic tin atom. For the Cu_2MnAl and Cu_2MnIn alloys, which have similar magnetic properties to the investigated alloy Cu_2MnSn, the nuclear magnetic resonance method[8] gave magnetic fields at the Mn and Cu nuclei which were of similar magnitude (about 200—236 kOe). Obviously, the same values of the field could be expected also for the Mn and Cu nuclei in the alloys we investigated. Hence, comparing the results reported in[8] and our measurements, we may draw the conclusion that the presence of considerable hyperfine fields at the nuclei of all the components of Heusler alloys agrees with the assumption[4] of the presence of a magnetic moment at copper atoms and makes it possible to apply the mechanism of an indirect superexchange via a nonmagnetic atom of tin to explain the nature of the ferromagnetism of these alloys. The values of the internal field obtained by us are close to the fields at the Sn^{119} nuclei in some ferrite garnets containing tin,[9] in which again the probable chief source of the field at the nucleus is assumed to be the polarization of the ion core of tin, due to an indirect superexchange. However, a detailed comparison of these results would be difficult because of the different nature of the chemical binding in these compounds.

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