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Temperature dependence of magnetic hyperfine fields in the metallic ferromagnets Rh_3FeSn and Rh_2CoSn

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Mössbauer γ spectroscopy is used to measure the temperature dependences of the magnetic hyperfine fields for ^{57}Fe and ^{119}Sn in the ordered metallic ferromagnets Rh_2FeSn and Rh_2CoSn. It is found that for Rh_2FeSn in the range $0.4 < T/T_c < 0.8$ the temperature dependences of the normalized fields $h(T) = H(T)/H(77\ K)$ are substantially different for the Fe and the Sn. To explain this temperature anomaly of the hyperfine field, a model is proposed in which account is taken of the temperature dependences of the local magnetization of the Fe and Rh atoms. The form of the temperature anomaly $h(T)$ for ^{119}Sn can be fully explained if account is taken of the fact that near T_c the temperature dependences of the magnetic moments of the Fe and Rh atoms are the same, but in the $0.4 < T/T_c < 0.8$ they are substantially different.

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1. INTRODUCTION

Among the unsolved problems of the theory of magnetic hyperfine interaction in metallic ferromagnets, a special role is played by the interpretation of the temperature dependence of the magnetic hyperfine fields. The great attention paid to this problem is due primarily to the attempts to obtain objective data on the nature of the hyperfine fields by explaining their temperature dependence. In particular, for nonmagnetic atoms (i.e., for atoms with zero intrinsic magnetic moments), the form of the temperature dependence of the hyperfine field may depend on the peculiarities of the mechanisms whereby the spin density is transferred from the magnetic environment to the nonmagnetic atom.

In a number of cases, nonmagnetic atoms reveal temperature anomalies of the hyperfine field, i.e., substantial deviations of the normalized hyperfine field $h(T) = H(T)/H(0)$ on the normalized magnetization of the system $M(T)/M(0)$. Particularly strong anomalies were observed for Sn impurity atoms in ferromagnetic metallic matrices. It is well known, for example, that Sn has a large anomaly in Fe (Refs. 1–3) and a giant anomaly in Co (Refs. 4 and 5). The reason why the anomalies are large precisely for the Sn atoms is qualitatively understandable. In this case, two large but oppositely directed contributions to the hyperfine field almost cancel each other [6] and therefore the absolute value of H should be very sensitive to small changes of the ratio of these contributions. Up to now, however, no unequivocal reason had been found for the temperature dependence of the relation between the two contributions. Several variants of an explanation of the anomalous behavior of $H(T)$ have been proposed for nonmagnetic atoms in metallic ferromagnets.[1-5,7] A common shortcoming of these explanations is that the initial premises are not very well founded and that the models cannot be quantitatively verified objectively.

As a rule, the experimental data are analyzed without allowance of the dependence of the hyperfine field on the thermal expansion of the crystal lattice; it was assumed that the corresponding corrections are negligible. Recently Möller[8] and Nikolaev *et al.*[9] obtained data on the dependence of H on the external pressure in the case of Sn. It follows from these results that in a number of cases the hyperfine field for Sn depends very strongly on the distance between the atoms. This means that the thermal expansion of the lattice can exert a strong in-

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fluence on the $H(T)$ dependence. Möller's data[8] were used by Price[3] to reconcile $H(T)$ with the constant-volume conditions for impurity Sn atoms in an Fe matrix. It was shown that the thermal expansion accounts for an appreciable share of the temperature anomaly of the hyperfine field.

If we use the more accurate data of Nikolaev *et al.*[9] then the $H(T)$ anomaly observed for Sn in Fe can be attributed to thermal expansion, at least up to temperatures on the order of 600 K. It is quite probable that the thermal expansion of the lattice is also the main cause of the giant anomaly of $H(T)$ for Sn in Co. It follows from the experimental data[9] that the change of the distance between the atoms influences greatly the negative contribution to the field, a contribution determined by the nearest neighbors of the Sn atom. If (as is the case for Sn in Co) the field at low temperatures is negative and very weak, one should expect large $H(T)$ anomalies due to thermal expansion of the lattice. It can therefore be assumed that it is precisely the thermal expansion which is one of the main causes of the large temperature anomalies of the hyperfine field for the Sn impurity atoms in the Fe and Co matrices. In any case, before we discuss other causes of these anomalies, the corresponding $H(T)$ dependences must be reduced to contant-volume conditions.

In this article we consider experimental data that point to existence of an $H(T)$ anomaly of another type. Mössbauer γ spectroscopy has shown that in the ordered ferromagnet Rh_2FeSn the $h(T)$ dependences are essentially different for ^{57}Fe and ^{119}Sn. As will be shown below, the temperature anomaly of the hyperfine field for the Sn atoms is in this case not connected with thermal expansion of the lattice, but can be attributed to the different temperature dependences of the logcal magnetizations of the Fe and Rh atoms.

2. EXPERIMENTAL TECHNIQUES AND FUNDAMENTAL RESULTS

The ordered ferromagnetic alloys Rh_2FeSn and Rh_2CoSn were recently synthesized by Suits,[10] who described their crystallographic and magnetic properties. The hyperfine interaction in these alloys for ^{57}Fe and ^{119}Sn at low temperatures was investigated by the Mössbauer spectroscopy method.[11,12] The magnetic hyperfine fields of ^{119}Sn at $T = 77$ K in Rh_2FeSn and Rh_2CoSn were found to be +57.3 and -54.9 kOe, respectively. Campbell *et al.*[12] obtained for these fields the values +55.1 and −51.1 kOe. The hyperfine field for ^{57}Fe in Rh_2FeSn is −288.5 kOe at 77 K. The Curie temperatures of the alloys are quite high, so that in the analysis of the results we shall assume that $h(77\text{ K})$ does not differ substantially from unity. We have used in this study alloys and resonant absorbers prepared by the same procedures as in the earlier studies.[10-12]

The Mössbauer resonant absorption spectra were measured with gamma sources in the form of $CaSnO_3$ or $BaSnO_3$ for ^{119}Sn and ^{57}Co in a copper matrix for ^{57}Fe. The temperature of the resonant absorber (in a cryostat or in an oven) was maintained constant with accuracy not worse than 0.01° with the aid of an electronic regulator. Some details connected with the procedure for the measurements and analysis of the spectra are given in a preceding paper.[11] The hyperfine-structure parameters for ^{119}Sn in Rh_2CoSn were measured in the range from 77 K to temperatures exceeding the Curie temperature, which was found to be $T_c = 444.1$ K (Ref. 13). In the case of Rh_2FeSn near $T_c = 583$ K (for both ^{57}Fe and ^{119}Sn) we observed strong broadening of the resonance lines, due apparently to spin-relaxation effects. In this connection, the hyperfine fields in Rh_2FeSn were determined in the 77–569 K range (corresponding to $T/T_c < 0.97$).

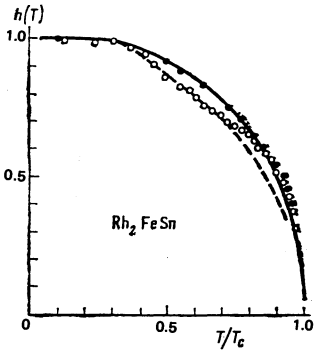

FIG. 1. Temperature dependences of the normalized hyperfine fields $h(T) = H(T)/H(77)$ in Rh_2FeSn: ●—for ^{57}Fe, ○—for ^{119}Sn. The statistical errors do not exceed the dimensions of the points. Solid—line Brillouin function $B_{3/2}$; dashed line—plot of $h(T)$ calculated for ^{119}Sn from formula (3).

The measured temperature dependences of the reduced hyperfine fields $h(T)$ are shown in Figs. 1 and 2. In Rh_2FeSn, the $h(T)$ dependence for ^{57}Fe agrees in almost the entire temperature range with the Brillouin function for $S = 3/2$; noticeable deviations from $B_{3/2}$ are observed only near T_c at $T/T_c > 9.85$. For ^{119}Sn in the same alloy, $h(T)$ at $T/T_c < 0.4$ and at $T/T_c > 0.8$ practically coincides with $h(T)$ for ^{57}Fe. In the intermediate temperature region ($0.4 < T/T_c < 0.8$) the $h(T)$ dependences for Sn and for Fe are substantially different: the plot of $h(T)$ for Sn exhibits a characteristic sag. The causes of this temperature anomaly of the hyperfine field in Sn will be considered below.

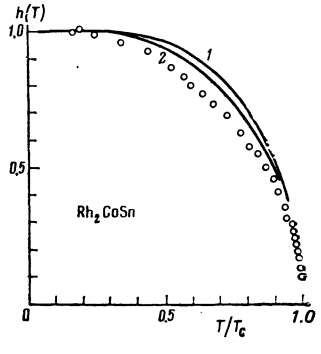

FIG. 2. Temperature dependence of normalized hyperfine field $h(T) = H(T)/H(77)$ for ^{119}Sn in Rh_2CoSn. The statistical error of each point does not exceed 0.02. Solid lines—Brillouin functions: 1—for $B_{1/2}$, 2—for B_1.

In the case of Rh_2CoSn, the $h(T)$ dependences for Sn (Fig. 2) has no clearly pronounced singularities, although it does not follow the Brillouin functions. [Figure 2 shows for comparison the functions $B_{1/2}$ and B_1, inasmuch as the magnetic moments of Co and Rh in Rh_2CoSn are close to $1\mu_B$ (Ref. 11)]. The results of the measurements of $h(T)$ for Sn in Rh_2CoSn in the critical temperature region were published by us earlier.[13]

3. TEMPERATURE ANOMALY OF THE HYPERFINE FIELD FOR Sn IN Rh_2FeSn

The compressibility and the coefficient of thermal expansion of the alloy Rh_2FeSn are unknown, so that it is impossible to evaluate quantitatively the effect of the thermal expansion on $h(T)$. It is obvious, however, that the obtained $h(T)$ anomaly for Sn in Rh_2FeSn is not connected with thermal expansion of the lattice. The correction for thermal expansion is a monotonic increasing function of the temperature,[3] therefore allowance for this correction can not explain the sag observed in the $h(T)$ dependence in the temperature range $0.4 < T/T_c < 0.8$. In addition, it follows from the data on the pressure dependence of H for Sn that the increase of the distance between the atoms is accompanied primarly by a decrease of the negative contribution to the field. The hyperfine field for Sn in the Rh_2FeSn alloy is positive, and therefore as a result of reducing the data to a constant volume the value of H for Sn should decrease and consequently the difference between the $h(T)$ dependences for Fe and Sn can only increase.

The anomalous form of the $h(T)$ dependence for Sn in Rh_2FeSn finds a natural explanation if account is taken of the possible difference between the temperature of the temperature dependences of the local magnetizations of the atoms Fe and Rh. The Sn atom has no intrinsic magnetic moment, so that the hyperfine field H at the nucleus of this atom is completely determined by the moments of the neighboring magnetic atoms. It was shown earlier[6] (see also Campbell *et al.*[12]) that the hyperfine fields measured at low temperatures for Sn atoms in metallic ferromagnets with bcc structure can be represented by the empirical formula

$$H = a\mu_1 + b\mu_2 + c\bar{\mu}, \tag{1}$$

where a, b, and c are empirical constants, μ_1 and μ_2 are the magnetic moments per atom in the first and second coordination spheres, respectively (relative to the Sn atoms), and $\bar{\mu}$ is the average atomic magnetic moment of the alloy. From an analysis of the experimental data it follows,[6,12] in particular, that for Sn the field H is a sum of two large contributions of opposite sign. The negative contribution is determined by the first term of formula (1), i.e., by the nearest neighbors of the Sn atom; the magnetic atoms in the more remote spheres determine the positive contribution, whose value is close to the absolute magnitude of the negative contribution. It is essential that the observed field H is as a rule much weaker than each of the two contributions, so that even a very small change of the ratio of the contributions should be accompanied by an appreciable change of H.

For metallic systems, formula (1) can be used also to analyze the temperature dependence of the field provided that under conditions of magnetic saturation the magnetic moments are replaced by their mean values at the given temperature:

$$H(T) = a\mu_1(T) + b\mu_2(T) + c\bar{\mu}(T). \tag{2}$$

We assume by the same token that the proportionality of each of the partial contributions of the corresponding local magnetization to H is preserved in the entire temperature range, and the proportionality coefficients, a, b, and c do not depend on the temperature.

This assumption leads directly to a simple and natural explanation of the experimental data. It is obvious that the anomalous form of the $H(T)$ function can arise if the system contains different magnetic atoms whose magnetic moments have different temperature dependences. This is precisely the situation in Rh_2F2Sn, where nonzero magnetic moments are possessed by the Fe and Rh atoms. The exact forms of the $\mu_{Fe}(T)$ and $\mu_{Rh}(T)$ relations are not known, and we shall therefore not discuss the results further, and use the molecular-field model. From an analysis of the magnetization and of the hyperfine fields, and also from the analogy with the properties of the ferromagnetic alloy FeRh, it follows[11,12] that the magnetic moments of the Rh and Fe atoms in Rh_2FeSn are close to $1\mu_B$ and $3\mu_B$, respectively. In the approximation of the model of the molecular field, the temperature dependences of these moment should follow the Brillouin functions $B_{1/2}$ and $B_{3/2}$. For the Fe atom this is directly confirmed by the experimental data: it follows from Fig. 1 that the relation $h(T)$ for Fe coincides with $B_{3/2}$ at $T/T_c < 0.85$. In this case the value of H for Fe is determined almost completely by the intrinsic moment of the atom, so that the functions $H(T)$ and $\mu_{Fe}(T)$ should be approximately proportional to each other.

We recognize that in Rh_2FeSn

$$\bar{\mu} = (2\mu_{Rh} + \mu_{Fe})/4,\ \mu_1(T) = \mu_{Rh}(T) = \mu_{Rh}(0)B_{1/2}(T),\ \mu_2(T) = \mu_{Fe}(T) = \mu_{Fe}(0)B_{1/2}(T),$$

and use the previously obtained values of the coefficients of formula (1) (Ref. 6) and the estimates of the atomic magnetic moments.[11] Then at $H(0) = +57.3$ kOe we get from (2), by elementary transformations,

$$h(T) = -1.3B_{1/2}(T) + 2.3B_{1/2}(T). \tag{3}$$

Since the functions $B_{1/2}$ and $B_{3/2}$ have different temperature dependences and the corresponding contributions to H are of opposite sign, one should expect an anomalous behavior of $h(T)$ in that temperature region where the Brillouin functions differ substantially from unity.

The $h(T)$ dependence for Sn, calculated from formula (3), is shown dashed in Fig. 1. It is seen that formula (3) accounts very well for the experimental data at $T/T_c < 0.7$. So good an agreement can be regarded to some degree as accidental, since the model based on the molecular-field theory is, naturally, approximate. Furthermore, the calculations do not take into account the

correction for the thermal expansion and neglect the tetragonal distortion of the Rh_2FeSn lattice. [This distortion can influence the values of the coefficients of formula (1).] It is nevertheless clear that the model considered above accounts at least qualitatively for the character of the anomaly of $h(T)$ for Sn at $T/T_c<0.7$.

It is easy to see why formula (3) does not agree with the experimental data at $T/T_c>0.7$. In the critical region of temperatures, the molecular-field model is certainly incorrect: in real ferromagnets, the behavior of the magnetization near T_c does not follow the Brillouin functions. The critical exponent β for the magnetization as given by the molecular-field model is 0.5, whereas the contemporary theoretical and experimental values of β for three-dimensional ferromagnets are close to 0.35–0.38. According to the universality principle, in the critical region the mean values of the local magnetization for the Fe and Rh atoms should have one and the same functional dependence on the temperature (regardless of their behavior at temperatures far from T_c). According to (2), such a temperature dependence should correspond also to the hyperfine field for the Sn atoms. This agrees well with our experimental data: at $T/T_c>0.8$ the $h(T)$ relations for Fe and Sn are the same but do not follow a Brillouin function.

Thus, the characteristic anomalous form of $h(T)$ for Sn in Rh_2FeSn can be fully explained by recognizing that the moments of Fe and Rh have identical temperature dependences near T_c, but substantially different ones in the range $0.4<T/T_c<0.8$. It appears that the temperature anomaly we obtained earlier[14] for the hyperfine field of impurity Sn atoms in the ferromagnetic matrix FeRh is explained in exactly the same way.

It is obvious that similar temperature anomalies of $H(T)$ should be observed also in other systems containing magnetic atoms with substantially different temperature dependences of the local magnetization. A well known example of systems in which the temperature dependences of the magnetization in nonequivalent lattice sites can be substantially different are certain ferrimagnetic compounds. Attention should be called in this connection to the result of Lyubutin *et al.*[15] who obtained a large $H(T)$ anomaly for impurity Sn atoms in the ferrite $MnFe_2O_4$. We note that when the magnetic hyperfine interaction in metallic and nonmetallic systems is considered it is customary to use entirely different concepts and approximations. In particular, no method has been found for representing the hyperfine fields in nonmetals with the aid of universal expressions similar to formula (1), so that the model considered above can not be used directly for an interpretation of the data obtained for nonmetallic ferrimagnets. Nonetheless, it is easy to note that there is an undisputed formal analogy between the model considered by us in the present article and the data interpretation contained in the paper of Lyubutin *et al.*[15]

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Kramers-Wannier transformation for systems with $Z(n)$ symmetry

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Duality relations analogous to the Kramers-Wannier symmetry of the two-dimensional Ising model are established for spin and gauge systems with internal symmetry $Z(n)$.

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Kramers and Wannier[1] drew attention to the fact that the two-dimensional Ising model possesses an exact symmetry that links the high- and low-temperature phases of this model. It is found that the model admits equivalent descriptions, in terms of a spin variable σ (the order parameter) defined on the lattice, and in

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