Hyperfine interactions for tin-119 impurity atoms in antiferromagnetic FeGe

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The hyperfine interactions of ^{119}Sn impurity atoms localized at the Fe sites in the hexagonal antiferromagnet FeGe is investigated by the Mössbauer spectroscopy technique. The magnetic hyperfine field H, quadrupole coupling constant, and isomer shift are measured between 4.5 and 730 K. A gigantic positive anomaly in the temperature dependence of $H(T)$ is observed: the hyperfine field increases with increase of temperature from 9.2 kOe at 4.5 K to 16.9 kOe at 230–290 K. The interrelationship between the temperature dependence of $H(T)$ and the thermal expansion of the crystal lattice is considered. The anomalous behavior of $H(T)$ is ascribed to the strong radial dependence of the partial contributions to the hyperfine field. The quadrupole coupling constant is negative and decreases linearly with increase of temperature. The asymmetry parameter of the electric field gradient is 0.85 ± 0.15. The strong temperature dependence of the resonance line shift is regarded as the result of the temperature dependence of the electron density in the vicinity of the Sn atomic nucleus. The asymmetry of the quadrupole doublet components in the paramagnetic temperature range can be attributed to the vibration anisotropy of the Sn atoms in the FeGe lattice.

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1. INTRODUCTION

One of the pressing problems of the physics of hyperfine interactions in solids is an explanation of the origin of the magnetic hyperfine fields H at nonmagnetic atoms in metallic magnets. It is customarily assumed that in this case the magnitude and sign of H are determined mainly by the Fermi contact interaction with the polarized conduction electrons. The theoretical analysis of the data on the magnetic hyperfine interaction is connected in this case with the general problem of the behavior of the conduction electrons in magnetic metals. Modern hyperfine-field models are qualitative in character, primarily because of the limited amount of actual information on the radial distribution of the spin density and on the singularities of the interaction of the conduction electrons with the potential of a nonmagnetic atom in a metallic ferromagnet or antiferromagnet.

Substantial progress was made in this field in recent years through the study of the hyperfine interaction of nonmagnetic atoms in ordered magnetic alloys and intermetallic compounds. These systems have a great variety of magnetic and structural properties, a fact that makes it possible, in particular, to investigate the dependence of the hyperfine field on the composition of the magnet and on the spatial distribution of the magnetic moments. Experiments of this type are particularly promising for impurity atoms, for in this case a comparative analysis of the results is possible for many impurity + matrix combinations. This method was used, for example, for the study[1] of the radial dependence of the partial contribution to H, and of the dependence of H on the charge of the nonmagnetic atoms[2] and on the distance between the atoms.[3]

In the present study we investigated, by Mössbauer γ spectroscopy, the hyperfine interactions for impurity ^{119}Sn atoms in the hexagonal antiferromagnet FeGe. Our main purpose was to study and interpret the temperature anomaly of the magnetic hyperfine field. In addition, data were obtained on the temperature dependences of the isomer shift, of the electric quadrupole interaction, and of the anisotropy of the vibrations of the impurity Sn atoms in the FeGe lattice.

The ordered hexagonal FeGe phase is produced by annealing the alloy at a temperature 650–700 °C.[4,5] The phase has a structure of type B35 (Fig. 1) with lattice constants $a = 5.00$ Å and $c = 4.05$ Å. According to neutron-diffraction data[5,6] hexagonal FeGe is a collinear antiferromagnet above 30.5 K; the magnetic moments of the Fe atoms (equal to 1.7 μ_B) are oriented parallel to the hexagonal axis of the crystal. In the basal plane, the Fe atoms form layers with ferromagnetic orientation of the moments. At temperatures below 30.5 K, a helicoidal magnetic structure is produced; the angle of deviation of the moments from the hexagonal axis increases with decreasing temperature and approaches 16° at 0 K.[5] The Néel temperature, according to data by various workers, is 398–412 K.[4,6-8] The magnetic hyperfine interaction for ^{57}Fe in FeGe was first investigated by Nikolaev et al.[7] A detailed study of the temperature dependences of the parameters of the hyperfine interaction for ^{57}Fe in hexagonal FeGe was recently made by Haggstrom et al.[8]

2. EXPERIMENTAL TECHNIQUE, ANALYSIS OF ABSORPTION SPECTRA, AND LOCALIZATION OF Sn ATOMS IN FeGe LATTICE

The samples were prepared by fusing the components (purity not worse than 99.8%) in high vacuum, tin en-

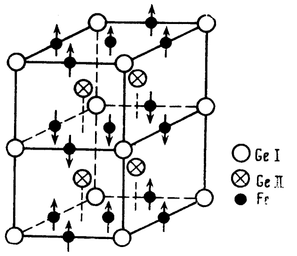

FIG. 1. Crystal and magnetic structures of hexagonal FeGe. The arrows show the orientation of the magnetic moments of the Fe atoms.

riched with ^{119}Sn to 91.3% was added to the iron beforehand. The ingots were remelted several times and then annealed in succession at 880 °C for 30 hours and at 650 °C for 100 hours. An x-ray analysis confirmed that an ordered FeGe phase with B35 structure was produced; the lattice constants agreed with the previously published data. As an additional check of sample quality, we measured the Mössbauer absorption spectra of the ^{57}Fe isotope; the results of these measurements agree well with the data of Haggstrom et al.[8]

Several alloys were prepared, with impurity tin concentration from 0.2 to 0.8 at.%; some of the samples deviated slightly from stoichiometry. The absorption spectra for ^{119}Sn at temperatures above 140 K were the same for all samples. For some samples with high tin concentration we observed at low temperatures, besides the lines of the main spectrum, also additional lines whose intensity depended very strongly on the temperature. The total intensity of these lines (for alloys with 0.8 at.% tin) at 77 K was ~10% of the total area of the spectrum, but at temperatures above 140 K they became practically unobservable. The cause of these lines remains unexplained, and they will not be discussed here. The Néel temperature T_N (which was determined from the temperature dependence of the hyperfine field for ^{110}Sn) decreased linearly with increasing tin concentration in accordance with the formula $T_N[\mathrm{K}]=399.4-15.6c$, where c is the tin concentration in at.%. Extrapolation to $c=0$ yields $T_N=399.4$ K, which agrees well, for example, with the value $T_N=398$ K obtained from the temperature dependence of the hyperfine field for ^{57}Fe in pure FeGe.[8] These results show that the impurity Sn atoms form a solid solution in the FeGe lattice.

The absorption spectra of the 23.9-keV Mössbauer gamma radiation of ^{119}Sn were measured with sources in the form of $\mathrm{CaSnO_3}$ or $\mathrm{BaSnO_3}$, using spectrometers with triangular dependence of the velocity on the time. We measured simultaneously the two identical (statistically independent) spectra; the results for two such spectra were averaged. For a thin $\mathrm{BaSnO_3}$ absorber the line width in the spectrum was 0.90 ± 0.02 mm/sec. The measurements were made in the temperature interval 4.5–730 K. The temperature of the samples (in cryostats or in the oven) was maintained constant with the aid of an electronic regulator with accuracy not worse than ±0.02 K. The absolute temperature was known accurate to 0.5 K.

For the Sn impurity atoms in FeGe, the energies of the magnetic dipole and electric quadrupole hyperfine interactions are comparable in magnitude. In this case it is necessary to use for the analysis of the spectra the total Hamiltonian of the hyperfine interaction. In a coordinate system whose axes coincide with the principal axis of the electric field gradient (EFG) tensor, the Hamiltonian is of the form

$$\mathscr{H}=-g\mu_N H[I_z\cos\theta+(I_x\cos\varphi+I_y\sin\varphi)\sin\theta]$$
$$+\frac{eQV_{zz}}{4I(2I-1)}[3I_z^2-I(I+1)+\eta(I_x^2-I_y^2)],\qquad(1)$$

where I is the spin of the nucleus, g is the nuclear g factor, μ_N is the nuclear magneton, e is the charge of the proton, Q is the quadrupole moment of the nucleus, $V_{zz}=\partial^2V/\partial z^2$ is the principal component of the EFG tensor, I_x, I_y, and I_z are the operators of the projections of the angular momentum, θ and φ are the polar and azimuthal angles and determine the orientation of the magnetic hyperfine field H. The components of the EFG tensor were chosen such that $|V_{zz}|\geqslant|V_{yy}|\geqslant|V_{xx}|$. In this case the EFG asymmetry parameter $\eta=(V_{xx}-V_{yy})/V_{zz}$ varies in the range $0\leqslant\eta\leqslant1$. For the ground state of the nucleus ($I=\frac{1}{2}$) the quadrupole interaction is equal to zero.

The eigenvalues and the eigenvectors of the Hamiltonian (1) for the excited ($I=\frac{3}{2}$) and ground ($I=\frac{1}{2}$) states of the nucleus were calculated with a computer as functions of the parameters θ, φ, η and $R=eQV_{zz}/2g\mu_N H$ (where g is the nuclear g-factor for the state $I=\frac{3}{2}$). In the general case the spectra contain eight resonance lines, whose energies (at zero isomer shift) are determined as the differences between the eigenvalues of the Hamiltonian for the two nuclear states. The components of the eigenvectors were used to calculate the relative intensities of the resonance lines. A preliminary estimate of the parameters θ, φ, η, and R for the experimental spectra was carried out with the aid of diagrams similar to the Kundig diagrams.[9] The ratio of the g factors of the ground and excited states of the ^{119}Sn nucleus was assumed to −4.602.

A computer reduction of the spectra by least squares yielded the hyperfine field H, the isomer shift, and the widths of the resonance lines. The spectra were measured with "thin" absorbers (not more than 1.0 mg/cm^2 in terms of the isotope ^{119}Sn), therefore each component of the spectrum was represented by a Lorentz function. In the reduction of certain spectra we introduced corrections that took into account the anisotropy of the oscillations of the Sn atoms (for details see below); allowance for these corrections at $T<T_N$ did not lead, however, to a substantial change of the results.

In connection with the high energy of the quadrupole interaction ($|R|>8$), the results of the spectrum reduction depended relatively little on the parameters R and η. The EFG asymmetry parameter was found to be $\eta=0.85\pm0.15$. The quadrupole-interaction constant was determined accurate to 0.02–0.04 mm/sec in the paramagnetic region of temperatures and 0.1–0.2 mm/sec in the antiferromagnetic region. The accuracy of the determination of H and of the isomer shift was respectively 0.2–0.5 kOe and 0.01–0.02 mm/sec. The widths of the hyperfine structure components ranged from 0.88 mm/sec (at high temperatures) to 1.14 mm/sec (at low temperatures); the increase of the width at low temperatures is due to the increase of the effective thickness of the absorber. An analysis of the spectra in the antiferromagnetic region ($T=4.5$–400 K) has shown that the quadrupole-interaction constant is negative ($R<0$) for the Sn atoms in FeGe. The best agreement between the theoretical spectra and the experimental ones was obtained at $\theta=0$, i. e., at parallel orientation of H and the Z axis of the EFG tensor. Figure 2 shows a comparison of the results of a computer calculation with one of

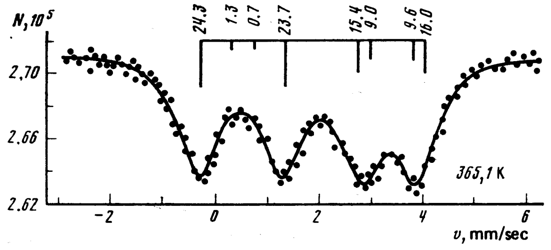

FIG. 2. Mössbauer-absorption spectrum for ^{119}Sn in FeGe, measured at $T = 365.1$ K. The theoretical curve (solid line) corresponds to the parameters $\theta = 0$, $\eta = 0.9$, $R = -11.4$, $H = 12.25$ kOe. The vertical lines show the positions of the components of the hyperfine structure; the relative intensities of the components are indicated by the numbers above the lines.

of the experimental spectra. The general character of the variation of the shape of the spectra as function of the temperature is shown schematically in Fig. 3.

The results of the measurements and of the analysis of the spectra have shown, in particular, the following: 1) At sufficiently low concentration, the impurity Sn atoms in the FeGe lattice occupy only one position 2) In the paramagnetic temperature region ($T > T_N$), an asymmetrical quadrupole doublet is observed in the spectra; it will be shown below that the asymmetry of the doublet is due to the anisotropy of the vibrations of the Sn atoms in the FeGe lattice. 3) The total splitting of (the distance between the outermost components of the hyperfine structure) first increases with decreasing temperature, reaching a maximum in the region $T = 230-290$ K, and then decreases, thus indicating an anomalous behavior of the temperature dependence of the hyperfine interaction. 4) At $T \geqslant 40$ K the orientation of the z axis of the EFG tensor coincides with the H direction ($\theta = 0$). The hyperfine field should have the same direction as the magnetic moments as the Fe atoms, consequently the z axis of the EFG tensor is parallel to the hexagonal axis of the crystal.

The obtained parameters of the quadrupole interaction make it possible to determine the character of the localization of the Sn atoms in the FeGe lattice. The Ge and Sn atoms have the same configuration of the external electrons. One can therefore expect the impurity Sn atoms to be localized at the Ge sites (as was the case with Sn impurity atoms in the Fe_3Ge lattice[3]). As seen from Fig. 1, the hyperfine field for the Ge II site should be identically equal to zero, since the total moments in each of its coordination spheres are equal to zero (just, for example, as for the Sn II site in the isostructural compound FeSn, Ref. 10). The surroundings of the two other sites, Fe and Ge I, have substantially different symmetries. For the Ge I site the surrounding has axial symmetry, and for the Sn atoms in this site the EFG tensor should also be axially symmetrical ($\eta = 0$). It was found by experiment that the parameter η is quite close to unity, corresponding to the strongly asymmetrical distribution of the charges around the Sn atom in the basal plane. This allows us to conclude that the impurity atoms Sn in the FeGe lattice are localized at the Fe site.

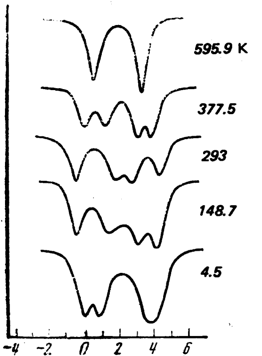

FIG. 3. Schematic representation of the absorption spectra for Sn in FeGe at various temperatures.

3. TEMPERATURE ANOMALY OF THE HYPERFINE FIELD FOR IMPURITY Sn ATOMS IN FeGe. CORRELATION WITH THE THERMAL EXPANSION OF THE LATTICE

The temperature dependence of the magnetic hyperfine field $H(T)$ for Sn impurity atoms in FeGe is shown in Fig. 4. For comparison, the same figure shows the temperature dependence of the hyperfine field for ^{57}Fe (Ref. 8), which corresponds to the temperature dependence of the average value of the magnetic moment of the Fe atoms. At $T = 4.5$ the field at the Sn is $H = 9.2 \pm 0.5$ kOe. In the range 4.5–60 K the field is almost constant; with further increase of the temperature, H is almost doubled for Sn, and reaches a maximum (16.9 ± 0.3 kOe) in the interval 230–290 K. The normalized hyperfine field $h(T) = H(T)/H(0)$ is traditionally compared with the normalized magnetization $m(T) = M(T)/M(0)$. In this representation, the function $H(T)$ for the Sn atoms is characterized by a gigantic positive anomaly of the quantity $h(T) - m(T)$, equal to 110–160% of $m(T)$ at temperatures 230–290 K. The considerable deviations of the function $h(T)$ from $m(T)$ are characteristic of Sn atoms in metallic magnets, but no positive anomaly of this size was previously observed.

The large anomalies of $H(T)$ for nonmagnetic atoms in metallic magnets are the subject of intense investigations and discussions; there is no meeting of minds concerning the cause of such anomalies. Even in the early stages of the investigations it was proposed[11 12] to consider the hyperfine field as a sum of proportional contributions of opposite sign with different temperature dependences. If each of these contributions is substantially larger than the summary observed field H, then even in small change of the ratio of the contribu-

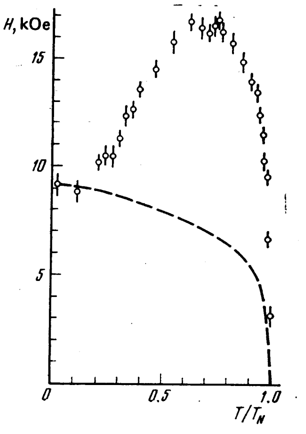

FIG. 4. Temperature dependence of the magnetic hyperfine field $H(T)$ for Sn in FeGe. The dashed curve shows the function $H(T)$ for ^{57}Fe (Ref. 8), normalized to the hyperfine field for Sn at $T = 4$ K.

tions could lead to large anomalies of $H(T)$. At the present time, the correctness of this assumption is almost obvious, but the question of the cause of the difference in the temperature dependences of the partial contributions remains open.

It was shown recently[13] that for impurity Sn atoms the $H(T)$ anomalies can be attributed to a strong dependence of H on the thermal expansion of the crystal lattice. In the general case, for a nonmagnetic atom in a metallic ferromagnet or antiferromagnet, the hyperfine field at $T=0$ K can be represented as a sum of contributions from all the surrounding magnetic atoms:

$$H(0)=\sum M_i(0)p(r_i^0), \tag{2}$$

where $M_i(0)$ is the magnetic moment of the i-th magnetic atom (in μ_B) at $T=0$ K, and $p(r_i^0)$ is the partial contribution to H for a moment equal to 1 μ_B and located at $T=0$ K at a distance r_i^0 from the nonmagnetic atom.

It is known that for the Sn atom the nearest magnetic neighbors determine the large contribution to the hyperfine field; the magnetic atoms located in more remote coordination spheres add up to a positive contribution of comparable magnitude.[1,3,13] It is obvious that the partial contributions p_i depend on the distances between the atoms and consequently on the thermal expansion of the lattice. An analysis of the dependence of H for Sn on the pressure[14] and a comparison of the hyperfine fields for the Sn atoms in isostructural matrices[3] have shown that a particularly strong dependence on the distance is a characteristic of the negative contribution to H; for this reason, when the distance between the atoms increases, one usually observes for Sn a decrease of the negative component of the hyperfine field. In a preceding paper[13] we explained the anomalies of $H(T)$ by using a somewhat modified procedure of reducing the data to a constant volume, and employed experimental data[14] on the dependence of H for Sn on the pressure. Unfortunately, suitable experimental data are still extremely scanty.

We consider below a new method of representing the results, which makes it possible to establish the existence of a correlation between the anomalies of $H(T)$ and the thermal expansion of the lattice in a very simple and lucid form. It is important that it is not necessary in this case to know the pressure dependence of H; nor is a concrete model representation of the nature of the hyperfine field used. We assume that the change of the distance between the atoms under thermal expansion is the only cause of the dependence of the partial contributions p_i on the temperature. If all the magnetic moments of the system have the same temperature dependence $m(T)=M_i(T)/M_i(0)$, then at the temperature T Eq. (2) takes the form

$$H(T)=\sum M_i(T)p(r_i^T)=m(T)\sum M_i(0)p(r_i^T), \tag{3}$$

where r_i^T is the distance to the i-th magnetic atom at the temperature T.

We consider now the quantity

$$\Delta H(T)=H(T)-H(0)m(T), \tag{4}$$

where $H(T)$ is the hyperfine field measured at the temperature T. The quantity $\Delta H(T)$ is thus the difference between the actually observed field and its ideal value $H(0)m(T)$, which would have been observed without thermal expansion. From (2)–(4) it follows that

$$\Delta H(T)=m(T)\left[\sum M_i(0)\frac{\partial p_i}{\partial r}\Delta r_i\right], \tag{5}$$

where $\Delta r_i=r_i^T-r_i^0$. (Since Δr_i are small, the derivatives $\partial p_i/\partial r$ can be regarded in the first-order approximation as constant and independent of temperature.)

The quantities Δr_i can be expressed in terms of the linear coefficient of the thermal expansion $\alpha(T)$. For a crystal with isotropic thermal expansion

$$\Delta r_i=r_i^0\int_0^T\alpha(T)dT=r_i^0 I(T),$$

where

$$I(T)=\int_0^T\alpha(T)dT$$

(generalization to the case of an anisotropic expansion can be carried out directly, but requires consideration of the concrete structure of the crystal.) Then Eq. (5) can be rewritten in the form

$$\Delta H(T)=m(T)I(T)\left[\sum M_i(0)\frac{\partial p_i}{\partial r}r_i^0\right]. \tag{6}$$

The sum in the square brackets does not depend on temperature; denoting this sum by C, we obtain the simple relation

$$\frac{\Delta H(T)}{m(T)}=CI(T). \tag{7}$$

If $\alpha(T)$ is known for this system, formula (7) can be easily compared with the experimental data, since the only unknown quantity is the proportionality constant C.

The essential difference between the new method of representing the experimental data and the traditional one is the absolute changes of the hyperfine field are considered rather than the relative ones. As seen from (6), $\Delta H(T)$ depends neither on the magnitude or the sign of the hyperfine field at $T=0$ K. The normalization of $H(T)$ to the hyperfine field at $T=0$ K does not correspond to the physical meaning of the problem, since $H(0)$ is determined by the values of the partial contributions p_i, while the change of the hyperfine field is determined by their derivatives $\partial p_i/\partial r$. According to (7), the temperature dependence of $\Delta H(T)/m(T)$ should have the same functional form as the thermal-expansion integral $I(T)$. This integral is usually positive, so that the sign of ΔH is determined by the sign of the proportionality constant C. If the model proposed by us is correct, the temperature dependence of $\Delta H(T)/m(T)$ for different systems should be similar to the same degree that the temperature dependence of $I(T)$ are similar for different substances. In particular, if the anomalies of $H(T)$ for the Sn atoms are determined primarily by the strong radial dependence of the negative contribution to the hyperfine field, then the functions $\Delta H(T)$ must be positive regardless of the sign of $H(0)$. The determination of the constant C may be of significant interest as a check on the theoretical models of the radial dependence of the partial contributions to the hyperfine field.

Figure 5 shows a comparison of Eq. (7) with the experimental data for impurity Sn atoms in an FeGe matrix (the present results) and in a matrix of cubic Co (the results of Cranshaw[11]). The anomaly of $H(T)$ for Sn and Co is the largest known anomaly of $H(T)$ of negative sign; thus, in the traditional representation these two anomalies have radically different forms.

In a new representation (Fig. 5), both relations are similar and have a functional form typical of the thermal-expansion integral $I(T)$. For Sn and Co, Eq. (7) agrees well with experiment in a wide range of temperatures and $C = 2.5 \times 10^3$ kOe. The coefficient of thermal expansion of FeGe was measured by Adelson and Austin[15]; unfortunately, the measurements were made only for temperatures above 298 K. We have calculated $I(T)$ for this case in the range 298–400 K and extrapolated the obtained dependence to the low-temperature region, assuming that $\alpha(T)$ for FeGe at low temperatures has no singularities whatever. This extrapolation is not completely unambiguous, but the resultant certainty cannot influence the main conclusion that a correlation exists between $\Delta H(T)$ and the thermal expansion of the lattice. The proportionality constant C for Sn and FeGe was found to be 8.7×10^3 kOe. In the 4–300 K range the relative increase of the distance between the atoms does not exceed 0.2%; the obtained large anomaly of $H(T)$ for Sn in FeGe points to a very strong dependence of the partial contributions p_i on the distance.

A quantitative interpretation of the obtained values of C is hardly possible at present, since it would require an explicit treatment of several partial contributions to the hyperfine field. For Sn in FeGe the values of these contributions, and all the more their derivatives, are unknown. The anisotropy of the thermal expansion of FeGe (Ref. 15) can limit somewhat the number of derivatives that make a substantial contribution to C, but for concrete conclusions we need more complete information on the partial contributions to the hyperfine field. We note that the dipole interaction cannot make a substantial contribution to the temperature anomaly of the hyperfine field, although the magnitude of the dipole contribution in this case can be comparable with the total value of $H(0)$. The dipole contribution to H is inversely proportional to the cube of the distance, and its value in this case does not exceed 10 kOe. Thus, in the range 0–300 K the change of H on account of the dipole contribution should be less than 0.1 kOe, which is negligibly small compared with the observed value of $\Delta H(T)$.

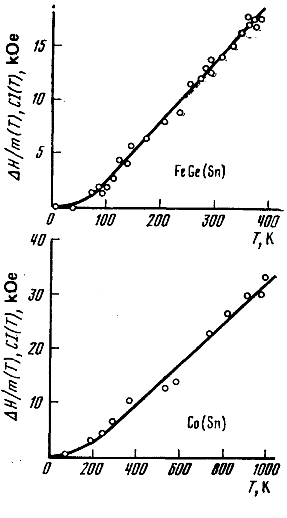

FIG. 5. Plots of $\Delta H(T)/m(T)$ for the impurity atoms Sn in an FeGe matrix (upper figure, results of present work) and in a matrix of cubic Co (lower figure, Cranshaw's results[11]). The solid lines show the thermal-expansion integrals $CI(T)$.

4. TEMPERATURE DEPENDENCES OF QUADRUPOLE INTERACTION, OF THE ISOMER SHIFT, AND OF THE ANISOTROPY OF THE VIBRATIONS OF THE Sn ATOMS IN THE FeGe LATTICE

The electric-field gradient in the region of the nucleus of the Sn atom in the FeGe matrix decreases linearly with increasing temperature. In the range 4.5–730 K the temperature dependence of the energy of the quadrupole interaction corresponds to the formula

$$\tfrac{1}{2}eQV_{zz}(1+\tfrac{1}{3}\eta^2)^{1/2}=\Delta(0)(1-aT), \tag{8}$$

where $\Delta(0) = 3.20 \pm 0.01$ mm/sec and $a = (2.58 \pm 0.06) \times 10^{-4}\,\mathrm{K}^{-1}$. It is known that for impurity atoms in metallic matrices the temperature dependence of the quadrupole interaction is frequently proportional to $T^{3/2}$ (see, e. g., Ref. 16). In our case, however, the linear relation (8) agrees much better with the experimental data.

An unexpected result was obtained for the temperature dependence of the shift of the resonance line. In the paramagnetic temperature region ($T \geq 400$ K) the shift was found to be a linear function of the temperature.

$$\delta(T)=a-bT, \tag{9}$$

where $a = 2.19 \pm 0.02$ mm/sec and $b = (5.2 \pm 0.4) \times 10^{-4}$ mm/sec × K. The experimental value of the coefficient b is much larger than its value for the thermal shift of the Mössbauer line in the classical limit ($b = 3.5 \times 10^{-4}$ mm/sec × K). Such a strong increase of the rate of change of the shift with changing temperature can apparently not be attributed to some singularities in the dynamics of the Sn atoms. We propose that this result means an explicit temperature dependence of the isomer shift, i. e., of the electron density in the region of the nucleus. In ordinary cases this contribution to the temperature shift of the Mössbauer line is as a rule negligibly small.

In the paramagnetic temperature region we have observed a strong dependence of the intensities of the quadrupole-doublet components (Fig. 3). The quadrupole-interaction constant for the atoms Sn and FeGe is negative, so that the line of minimum intensity corresponds to a transition from the level $I_z = \pm\frac{3}{2}$. The asymmetry of the components of the quadrupole doublet can be due to two causes: the anisotropy of the vibrations of the Mössbauer atom (the Gol'danskiĭ-Karyagin effect) and to the texture of the resonant absorber.

To choose between these two possibilities, we measured the dependences of the asymmetry of the doublet

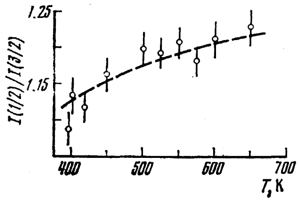

FIG. 6. Temperature dependence of the asymmetry of the quadrupole doublet $I(\frac{1}{2})/I(\frac{3}{2})$ for ^{119}Sn in FeGe. The dashed line shows this dependence calculated in the Debye approximation for the case of axial symmetry of the amplitudes of the tin-atom vibrations.

on the temperature and on the orientation of the absorber relative to the direction of the gamma-quantum flux. The temperature dependence of the intensity ratio $I(\frac{1}{2})/I(\frac{3}{2})$ is shown in Fig. 6. It is seen that the asymmetry increases with increasing temperature, a characteristic feature of the Gol'danskiĭ-Karyagin effect. By using the "magic angle" method,[17] we have shown that the influence of the texture (of the predominant orientation of the crystallites) on the asymmetry of the doublet is negligibly small. The results of these two experiments allow us to conclude that for tin in an FeGe lattice the asymmetry of the quadrupole doublet is due to the anisotropy of the vibrations of the Sn atoms.

It was shown above that distribution of the electron density around the Sn atom is characterized by a large EFG asymmetry parameter. It is quite probable that in this case the tensor of the amplitudes of the vibrations of the Sn atoms may likewise not be axially symmetrical. The probability of the Mössbauer effect depends in this case on both the polar and the azimuthal angle, and the principal axes of the vibration tensor need not necessarily coincide with the principal axis of the EFG tensor. An analysis of the Gol'danskiĭ-Karyagin effect for this general case[18] becomes ambiguous because of the large number of free parameters, which are difficult to estimate without additional information on the singularities of the dynamics of the atoms. For this reason we have confined ourselves to an approximate analysis of the results, and consider a variant with axial symmetry of the atom vibrations ($\langle x^2\rangle = \langle y^2\rangle \neq \langle z^2\rangle$) in the Debye approximation.

The average effective Debye temperature $\langle \Theta_D\rangle$ was determined from a temperature dependence of the total intensity of the resonant absorption; in the 400–730 K range this dependence corresponds to $\langle \Theta_D\rangle = 230 \pm 10$ K. Using this value of $\langle \Theta_D\rangle$ and results of the measurements of the symmetry of the quadrupole doublet, we can calculate the temperature dependences of the mean squared vibration amplitudes $\langle x^2\rangle$ and $\langle z^2\rangle$. In the Debye approximation these dependences correspond to effective Debye temperatures $\Theta_z = 175 \pm 7$ K for vibrations along the hexagonal axis and $\Theta_x = 286 \pm 17$ K in the basal plane. Thus, in our approximation the amplitude of the vibrations of the atom along the hexagonal axis is much larger than the basal plane. For example, at $T = 500°$ K the ratio is $\langle z^2\rangle/\langle x^2\rangle = 2.5 \pm 0.5$. The temperature dependence of the asymmetry of the quadrupole doublet, calculated in the Debye approximation, is shown in Fig. 6.

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