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Abstract

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PHYSICS

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THE EFFECT OF THE EXCHANGE INTERACTION OF MAGNETIC IONS ON THE SHIFT OF THE MÖSSBAUER SPECTRUM

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The energy of the exchange interaction of magnetic ions in a crystal depends on the distance between the interacting ions. In crystals with a high concentration of magnetic ions, exchange interactions can contribute to the elastic constants of the crystal lattice. Since the relativistic shift of the Mössbauer spectrum depends on quantities characterizing the vibrational spectrum of the crystal ⁽¹⁾, exchange interactions apparently can influence the shift of the center of gravity of the Mössbauer spectrum δE . Since the energy of exchange interaction also depends on the relative orientation of the spins of the interacting ions, the shift of the Mössbauer spectrum must change when the degree of ordering of the spin system of the crystal changes. The latter is the most significant point, since it is precisely this that makes it possible to observe the phenomenon under discussion.

A theoretical treatment of this phenomenon, carried out in the harmonic approximation on the basis of the general theory of the relativistic shift ⁽²⁾, showed that the presence of exchange interaction leads to an additional shift of the center of gravity of the Mössbauer spectrum ^(3,4).

On the basis of the Debye model of the phonon spectrum of a crystal, taking into account the exchange interaction only of nearest neighbors, we obtained a comparatively simple relation ^(3,4), which showed that the exchange shift δE^{ex} depends on the product

$$A = \langle \hat{S}_i \cdot \hat{S}_j \rangle \cdot I'',$$

where $\langle \hat{S}_i \cdot \hat{S}_j \rangle$ is the mean value of the scalar product of the spin operators of the interacting magnetic ions, and I'' is the second derivative of the exchange integral with respect to the spatial variables; moreover, $\delta E^{\text{ex}} = 0$ if $A = 0$. As a result, in the paramagnetic state the exchange shift is absent, since $\langle \hat{S}_i \cdot \hat{S}_j \rangle = 0$. In the ferromagnetic ($\langle \hat{S}_i \cdot \hat{S}_j \rangle \approx S^2$) and antiferromagnetic ($\langle \hat{S}_i \cdot \hat{S}_j \rangle \approx -S^2$)

Figure 1

Figure 1: Figure 1

states the exchange shift has one and the same sign, since the signs of I'' for ferro- and antiferromagnets are opposite.

Thus, upon transition from the paramagnetic to the ferro- or antiferromagnetic state, a jump-like change in the shift should occur, associated with a change in the value of $\langle \hat{S}_i \cdot \hat{S}_j \rangle$. According to our estimates ^(3,4), the jump in the shift should be of the order of $0.1 \div 0.01$ mm/sec (Fe^{57}). A change in the shift upon passage through the Curie point T_C (the Néel point T_N) has been observed in metallic iron ⁽¹⁾, in nickel on impurity nuclei Fe^{57} ⁽⁵⁾, in the intermetallic compound FeSn (on Fe^{57}) ⁽⁶⁾, and elsewhere ^(10–12).

Various assumptions have been advanced concerning the possible causes of the jump-like change in the shift at the transition points T_C or T_N ⁽⁷⁾, not, however, supported by calculations. The experiment described in the present article, as we believe, confirms the viewpoint set forth above.

The idea of the experiment consists in comparing the temperature dependence of the shifts of the Mössbauer spectra of two crystals which, in a rough approximation, differ only in the magnitude of the exchange interaction of the magnetic ions, with one of the crystals having the transition point T_C or T_N in the temperature interval under consideration. The magnitude of the shift for these samples should differ by an amount approximately equal to δE^{ex} in the temperature range below T_C (T_N). In the temperature range above T_C (T_N), the shifts should have substantially closer values.

Fig. 1. Temperature dependence of the shift δE of the Mössbauer spectrum of Fe^{3+} in $\alpha\text{-Fe}_2\text{O}_3$ (1) and in $\alpha\text{-Ga}_2\text{O}_3$ (2). The values of δE are given relative to the line of a Co^{57} source in stainless steel.

We investigated polycrystalline samples of $\alpha\text{-Fe}_2\text{O}_3$ ($T_N = 956^\circ\text{K}$) and $\alpha\text{-Ga}_2\text{O}_3$ with an admixture of Fe^{57} isomorphously replacing gallium ions (impurity content about 15%), in the temperature interval from 80 to 1080°K.

Crystals of $\alpha\text{-Fe}_2\text{O}_3$ and $\alpha\text{-Ga}_2\text{O}_3$ crystallize in the trigonal system of the corundum type (space group $R\bar{3}c$). The parameters of the crystal lattices are approximately the same ⁽⁹⁾. The mass numbers for Fe^{57} (57) and Ga (69) are comparatively close. The close similarity of the crystals makes it possible to assume that, if exchange interaction is excluded from consideration, the phonon spectra of the two indicated crystals will be similar.

The experimental data are presented in Fig. 1, which shows the dependence of the shift δE on temperature (the temperature dependence of the shift in $\alpha\text{-Fe}_2\text{O}_3$ was also studied in work ⁽⁸⁾ in the temperature interval from 80 to 900°K). The shift values for Fe^{3+} in $\alpha\text{-Fe}_2\text{O}_3$ and $\alpha\text{-Ga}_2\text{O}_3$ in the temperature

interval below $T_N = 956^\circ\text{K}$ differ by an amount of the order of $0.05 \div 0.06$ mm/sec. In the region of T_N , the spectrum shift of $\alpha\text{-Fe}_2\text{O}_3$ undergoes a jump of the order of 0.05 ± 0.02 mm/sec, and at temperatures above T_N the shifts of $\alpha\text{-Ga}_2\text{O}_3(\text{Fe})$ and $\alpha\text{-Fe}_2\text{O}_3$ coincide.

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