

MAGNETIC RESONANCE IN DIAMAGNETICALLY DILUTED POTASSIUM FERRITE SINGLE CRYSTALS

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UDC 548:537.611.46

Potassium ferrite ($\text{KFe}_{11}\text{O}_{17}$) possesses the hexagonal structure of β -alumina ($\beta\text{-Al}_2\text{O}_3$) belonging to the D_{6h}^4 spatial symmetry group. The unit cell of potassium ferrite comprises two four-layer spinel blocks divided by a potassium-oxygen layer. Ferrites of this structure are characterized by increased mobility of monovalent ions in the dividing (conducting) layers caused by the fact that about one third of interstices are localized in these layers. Therefore, depending on the conditions of synthesizing, deviations from the stoichiometry are quite probable for these compounds, for example, in the form of excessive monovalent K^+ ions or oxygen interstitial defects. Due to this, potassium ferrite is a semiconductor with electron and ion conductivity. In due time this circumstance determined interest in the study of the electric properties of this compound that can be used as an electrode-electrolyte pair of solid-state batteries. From practical viewpoint, $\text{KFe}_{11}\text{O}_{17}$ is interesting also as an antiferromagnet with high ordering temperature ($T_N = 800$ K [1]) and hence as a potential material for electronics of millimeter and submillimeter wavelengths.

In due time, Gorter [1] proposed a model of the magnetic potassium ferrite structure comprising antiferromagnetically ordered ferrimagnetic spinel blocks. It was assumed that the antiferromagnetic bond is formed by indirect exchange through the potassium-oxygen layers. In the present study, an attempt is made of testing the adequacy of this model by means of investigation of the influence of nonstoichiometry and diamagnetic substitution of Fe^{3+} ions in potassium ferrite on the mechanisms of exchange through the potassium-oxygen layers.

The $\text{K}_{1-y}\text{Fe}_{11-x}\text{Ga}_x\text{O}_{17}$ ($x = 0, 0.85, \text{ and } 1.55$ and $y = 0.35\text{--}0.62$) single crystals were grown by the method of spontaneous crystallization from the solution-melt using the technology described in [2]. The frequency-field, orientation, and temperature dependences of the resonant absorption in the potassium ferrite crystals were investigated with a broadband antiferromagnetic resonance spectrometer in pulsed magnetic fields up to 100 kOe for frequencies in the range 9–180 GHz and temperatures in the interval 4.2–300 K.

Two resonant absorption lines were detected; one of which was attributed to the antiferromagnetic resonance of the easy-plane antiferromagnet, and another was attributed to the resonance of isolated ions or clusters formed by the Fe^{3+} ions. The intensity of the second resonance for different samples was several ten times less than the antiferromagnetic resonance intensity and differed significantly from sample to sample. It was revealed that short-term sample annealing in air (for ~1 h at $T = 700^\circ\text{C}$) resulted in disappearance of the second resonance, while longer-time annealing of the same samples (for ~3–6 h) in the nitrogen vapor atmosphere restored the given resonance. The resonance of impurity nature can be caused by crystals of oxygen and by formation of electron holes that influence the exchange bonds through the oxygen ions located in definite positions. According to the Mössbauer data [3] obtained in the study of these crystals, the reason for the impurity resonance can be an excess in the K^+ ions which for different samples could be $y \sim 0.35\text{--}0.62$. According to the same data, in diamagnetically substituted potassium crystals, the Fe^{3+} ions are substituted by the Ga^{3+} ions primarily in the tetrahedral $4f$ -positions bounded with the potassium-oxygen layers through which the indirect exchange between spinel layers occurs.

We have established that with increase in the gallium concentration, the temperature of transition into the ordered state of crystals with $x = 0.85$ decreased to ~200 K, and in crystals with $x = 1.55$ it decreased to ~20 K. A comparison of these data with the Mössbauer data presented in [3] gives us grounds to believe that the observation of the resonance of impurity nature and of the strong dependence of the Néel temperature on the gallium concentration in the examined crystals are most likely caused by diamagnetic switch off of the strongest exchange bonds between the spinel blocks. A comparison

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of our data with the Mössbauer data presented in [3] also indicates that the Gorter model of the magnetic potassium ferrite structure has the right to existence. However, the final conclusion can be made only after static magnetic measurements with gallium-substituted potassium ferrite crystals.

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