Quantum jumps in the magnetization of a magnet with antiferromagnetic impurities

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The differential magnetic susceptibility of $Gd_xY_{3-x}Fe_5O_{12}$ ($0.01<x<0.2$) polycrystalline compounds is studied in the temperature range 0.6 to 4.2 K and in pulsed magnetic fields up to 50 T. The magnetization of the compound with the lowest Gd content ($x=0.01$) shows instead of a classical, a quantum (stepwise) phase transition to a completely saturated ferromagnetic phase at $T=0.6$ K, in magnetic fields around 33 T. To explain these results, a Heisenberg ferrimagnetic model with weakly coupled antiferromagnetic impurities is used. Following this model, the nonequidistance of the energy levels of the impurity ions causes the occurrence of susceptibility oscillations under a variation of external magnetic fields.

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INTRODUCTION

Diluted magnetic impurities embedded in a crystal matrix can play an important role as a local sensor, giving information about the intrinsic magnetic features of the matrix. The impurity alters the spectrum of the magnetic excitations, leading to the addition of local or quasilocal energy levels due to the interactions between impurity and matrix. As a result, the study of these levels can give detailed information about these magnetic interactions.[1,2] The presence of impurities moreover can drastically change the thermodynamic properties of the matrix, causing unusual field and temperature dependencies of the magnetization, magnetic resonance and specific heat.[3–5]

In the early 1970s another type of macroscopic quantum phenomena was predicted giving rise to field induced oscillations in the specific heat and susceptibility of materials with a ferromagnetic matrix and antiferromagnetic impurities.[6,7] These oscillations are the direct consequence of the magnon-magnon interaction between the matrix and the impurity,[6] which causes an unequal spacing of the $2S_0+1$ impurity energy levels where S_0 denotes the impurity spin. It is important to note that such oscillations represent another type of non-cooperative quantum phenomena, because mutual interaction between the impurities leads to a quick broadening of the impurity energy levels and can suppress all anomalies of the thermodynamic properties.[7]

The first attempt to discover these oscillations was made shortly after the theoretical prediction. The differential magnetic susceptibility of $Gd_xY_{3-x}Fe_5O_{12}$ ($x<0.2$) polycrystalline samples was studied at $T<5$ K in pulsed magnetic fields up to 50 T.[8] No evidence of oscillations was observed even at the lowest impurity content ($x=0.01$) and temperature (1.6 K). On the other hand, the experiments did not reveal the classical result for low impurity content ($x\leqslant 0.05$), because in that case the field interval ΔH, where the spin reversal takes place, does not depend on the impurity content. Moreover, for $x=0.01$ the value of ΔH is five times larger than predicted by the classical approach.[8] This result contradicts the classical molecular-field theory, but is in accord with the microscopic model.[6,7]

In this work we present the direct observation of field induced oscillations of the impurity susceptibility in a ferrimagnetic matrix. The main aim of this work is to demonstrate that under certain conditions, as predicted in Refs. 6 and 7, these oscillations appear due to quantum transitions between discrete $2S_0+1$ energy levels of the impurity spin. Our paper deals with the investigation of the differential magnetic susceptibility (χ) of $Gd_xY_{3-x}Fe_5O_{12}$ ferrites at very low temperatures (down to 0.6 K) in pulsed magnetic fields (up to 50 T). For $x=0.01$ the splitting of the χ peak in to two peaks is evident when temperature is decreasing and points to the quantum character of the Gd impurity magnetization process.

EXPERIMENTAL RESULTS

The differential magnetic susceptibility of $Gd_xY_{3-x}Fe_5O_{12}$ ($x=0.2;0.01$) was studied in pulsed magnetic fields up to $\mu_0 H=50$ T in the temperature range $0.6<T<4.2$ K. The $Gd_xY_{3-x}Fe_5O_{12}$ ferrite-garnets were the same as used in Ref. 8 and were prepared by standard ceramic technology from high purity oxides (rare earth impurity content $<10^{-3}$%). In order to obtain homogeneous material, the ingots were crumbled to a fine powder and then remelted up to five times. Cylindrical ingots were 15 mm in length and 5 mm in diameter. The impurity content was measured after the final annealing of the ingots by x-ray luminescence analysis and mass-spectrometer analysis.

Pulsed magnetic fields were generated by the discharge of a condenser bank into a wire-wound multiturn coil; the duration of the field pulse was about 26 ms.[9] The susceptibility was measured using a pulsed inductive magnetometer of an ''open'' type.[10] Analog to digital transformation of the dM/dt and dH/dt signals was carried out by means of a Bakker 256 transient recorder with 12-bit resolution at 300 kHz. The sensitivity of the inductive sensor to a magnetic moment was around 10^{-2} emu in fields above 30 T.

It is well established[8] that the magnetic structure of $Gd_xY_{3-x}Fe_5O_{12}$ in an external field up to 50 T can be described in the terms of a two-sublattice model. One sublattice forms the Fe^{3+} ions and its magnetic moment is $\mathbf{M}_{Fe}=5\mu_B$ per formula unit. The other sublattice is formed by the Gd^{3+} ions and has a magnetic moment $\mathbf{M}_{Gd}=(7x)\mu_B$ per formula unit where x is concentration of the impurity. The antiferromagnetic exchange interaction between these two magnetic

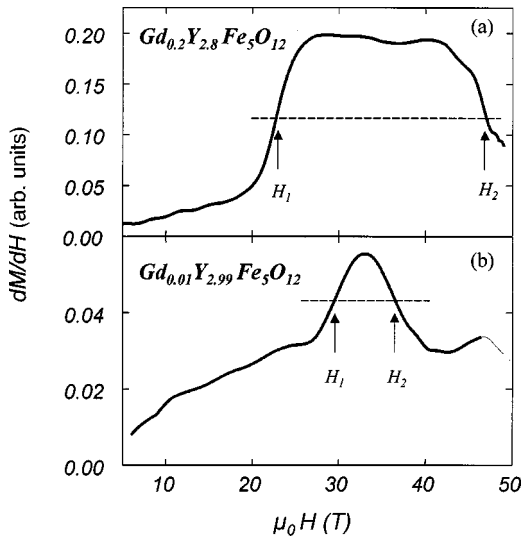

FIG. 1. Field dependence of the differential magnetic susceptibility for $Gd_xY_{3-x}Fe_5O_{12}$ samples at $T=4.2$ K with different Gd concentrations: $x=0.2$ (a) and $x=0.01$ (b).

subsystems leads to antiparallel alignment of $\mathbf{M}_{\mathrm{Fe}}$ and $\mathbf{M}_{\mathrm{Gd}}$ at $H=0$, and $|\mathbf{M}_{\mathrm{Gd}}|<|\mathbf{M}_{\mathrm{Fe}}|$ for $x<0.6$.[11] According to classical approach,[11,12] the application of an external magnetic field causes the reversal of the $\mathbf{M}_{\mathrm{Gd}}$ in the field $H_1<H<H_2$, where $H_{2,1}=H_{\mathrm{ex}}[1\pm M_{\mathrm{Gd}}/M_{\mathrm{Fe}}]$—here H_{ex} is the effective field of the Fe-Gd exchange interaction. As a result, within the classical model, the rotation of both moments takes place in this field interval and the total magnetic moment $M=\mathbf{M}_{\mathrm{Fe}}+\mathbf{M}_{\mathrm{Gd}}$ is practically linear. So, there are no oscillations of susceptibility in the field interval $\Delta H=H_2-H_1$.

A first set of measurements was carried out for both samples ($x=0.2;0.01$) at $T=4.2$ K in order to reproduce the results reported in Ref. 8. The field dependences of the differential magnetic susceptibility $\chi=dM/dH$ is presented in Fig. 1 for both samples. These curves are used to determine the values of the critical fields H_1 and H_2, which delimit fields, where the reversal of the impurity magnetic moments occurs. The field values at half-height of the susceptibility peak were taken to define H_1 and H_2 (see Fig. 1). It is well known that in fields at low temperature in field $H<H_1$ and $H_2>H_2$ the garnet susceptibility is small (less or around 10^{-4}) and slightly depends on magnetic field.[12] In these two cases collinear magnetic structure exist: ferromagnetic below H_1 and ferrimagnetic above H_2,[11] and magnetic susceptibility is determined only by paraprocesses inside the rare earth and the iron subsystems.[12] Within the field range $H_1<H<H_2$ the susceptibility is much larger due to appearance of noncollinear magnetic structure of the magnetic moments $\mathbf{M}_{\mathrm{Fe}}$ and $\mathbf{M}_{\mathrm{Gd}}$, and has to be constant, when the temperature broadening is neglected. The area, where a noncollinear phase exists, is much smaller for the $x=0.01$ sample, due to the reduction of the Gd content. At the same time, the observed ΔH value for this compound is five times higher than the value predicted by the classical model.[11,12]

The fields H_1 and H_2, derived from our experiments, are plotted in the field-temperature phase diagram of $Gd_xY_{3-x}Fe_5O_{12}$ for $x=0.01$ and $x=0.2$ together with the

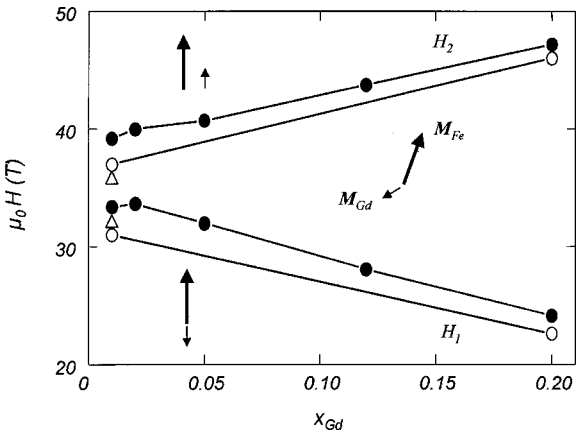

FIG. 2. Characteristic fields $H1$ and $H2$ of $Gd_xY_{3-x}Fe_5O_{12}$ versus the Gd content at $x\leqslant 0.2$. Solid circles are the results from Ref. 8 at $T=4.2$ K; open circles ($T=4.2$ K) and triangles ($T=0.6$ K) are the results of this work. The arrows demonstrate schematically the magnetic structure in different fields.

data from Ref. 8 (Fig. 2). Both data sets coincide well and the small discrepancy ($\approx 5\%$) can be attributed to the difference in field calibration.

Figure 3 shows the field dependences of the differential magnetic susceptibility for the $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ compound, measured at three different temperatures. The temperature decrease leads to a reduction of ΔH, but its value at $T=0.6$ K is still much larger than predicted by the classical model (~ 1 T). This is an essential deviation from the classical approach, as noted in Ref. 8. Moreover, the temperature produces substantial changes in the susceptibility of the mixed garnet: the initial broad peak begins to split at $T=1.5$ K and there are already two well defined peaks at $T=0.6$ K. Small anomalies of the dM/dH at this temperature in field below 29 T and above 38 are not reproducible (in contrast with two central peaks). This is the noise caused by mechanical vibration of the measuring cell, and it becomes larger with the temperature decrease. To improve the signal/noise ration, the data obtained at $T=0.6$ K were averaged

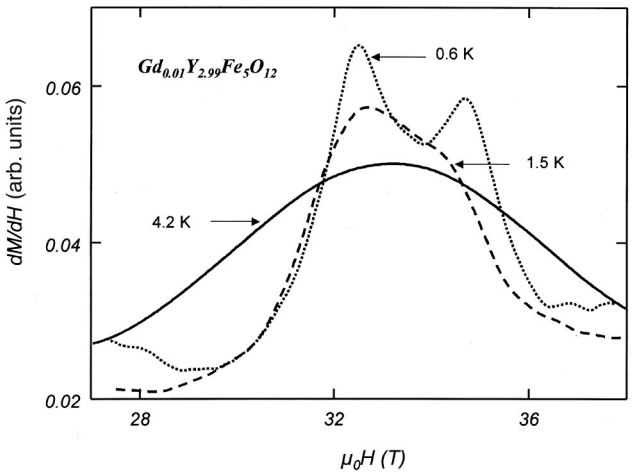

FIG. 3. Field dependences of the differential magnetic susceptibility of a $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ sample at different temperatures.

over seven pulses in contrast with $T=4.2\,\mathrm{K}$, when this procedure was done only over two pulses of magnetic field.

The observed splitting of the susceptibility peak for the $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ sample at $T=0.6\,\mathrm{K}$ points to the appearance of a qualitatively different, purely quantum, feature in the magnetization: the stepwise behavior as predicted in Refs. 6 and 7. The next section is devoted to numerical simulations, based on the models described in these two publications. We will start with the simplest model,[6] which demonstrates well the physical mechanism of the susceptibility oscillations in a magnet with antiferromagnetic impurities. After that more complicated calculations will be presented, based on the advanced model.[7] These simulations show qualitative agreement with our experiments, confirming the quantum phenomenon.

DISCUSSION

Let us consider a two-sublattice model for a garnet with nearest-neighbor interaction in a cubic lattice. The two sublattices (with the spins S_1 and S_2) alternate in such a way that the nearest neighbors always interact with the other sublattice. The impurity spins S_0 are weakly coupled with the matrix $|J_0| \ll |J|$, where $J>0$ is the exchange integral between the matrix spins S_1 and S_2, coupled antiferromagnetically and J_0 is the exchange integral between S_0 and S_1 or S_2 (these matrix spins are also coupled antiferromagnetically with the impurity spin). The impurity concentration is so small that one can consider all impurity ions as noninteracting; the temperature is assumed to be low compared with the Curie point (T_c) of the pure material ($T \ll T_c \propto J$, but the relation between T and J_0 can be arbitrary). The spin-wave approximation that was used in Ref. 6 to describe the matrix allows only small deviations of S_1 and S_2 from full saturation. The deviations from the impurity Néel state (where $M_0=-S_0$), however, can be large. The impurity states are described exactly, but the interaction of a well-localized impurity spin with the boson field of the matrix magnons is solved using standard perturbation theory.

According to Ref. 6, the solution of the Schrödinger equation at $J_0>0$ shows that there is no damping of the impurity states and that the impurity energy levels are given by

$$\varepsilon_m = m(g_0\mu_B H-\gamma)+\frac{1}{2}m(m-1)\alpha, \tag{1a}$$

where

$$\gamma = zSJ_0+S_0\alpha. \tag{1b}$$

and

$$\alpha = z(J_0)^2(I_W-1)/J. \tag{1c}$$

Here z is the number of nearest neighbors, g_0 is g factor of the impurity ion, I_W is the Watson integral (which is equal to 1.515, 1.393, and 1.345 for the simple cubic, bcc, and fcc lattices, respectively), $S=S_2-S_1$ and $m=S_0-M_0$. The term α describes the magnitude of the magnon-magnon interaction between the matrix and the impurity.

In spite of the fact that the model is developed under the approximation that the impurities substitute the matrix ions, this approach seems to be applicable in cases when impurity ions are located in interstitial positions, like Gd^{3+} ions in $Gd_xY_{3-x}Fe_5O_{12}$. This is one of the results of Ref. 6, where it was found that only long-wavelength magnons with $\varepsilon \leqslant zS_{1,2}J_0$ are significant for the magnon-magnon interaction between the matrix and impurity spins. For such long-wave magnons, the short-range order in the magnetic unit cell becomes unimportant, because all phenomena are determined by the averaged characteristics of the unit cell. In this case the ferrite matrix becomes equivalent to a ferromagnet with total spin $S=S_2-S_1$, and real location of the impurity spins does not play an important role.

One can see from Eq. (1a) that the energy levels are not equally spaced if α is nonzero. The difference between the ground level ($m=2S_0$) and the first excited level ($m=2S_0-1$) is smaller then that between the first and second excited levels, and so on. As a result, by increasing the magnetic field, the magnetization of the impurity spins becomes steplike and the differential magnetic susceptibility exhibits peaks.

The free energy of a matrix with an ideal gas of noninteracting impurities can be described as follows:

$$F=-\frac{1}{2}zJS^2+xzJ_0SS_0-gH\mu_BS-g_0\mu_BHS_0x + F_{sw}+F_{im}, \tag{2a}$$

where

$$F_{sw}=T\int_{gH}^{\varepsilon_{\max}}\ln(1-e^{-\beta\varepsilon})\rho(\varepsilon)d\varepsilon, \quad \beta=1/kT,$$

$$F_{im}=-xT\ln\sum_{m=0}^{2S_0}e^{-\beta\varepsilon_m}. \tag{2b}$$

Here the ferrite-garnet is considered as ferromagnet with total spin $S=S_1-S_2$. The first four terms in Eq. (2a) correspond with the free energy of the unperturbed initial states of the matrix and the impurity spins. The term F_{sw} in Eq. (2a) describes the contribution of the spin waves (magnons of the continuous spectrum), $\rho(\varepsilon)$ is the energy density of spin wave states in the presence of impurities.[3] The last term in this equation shows the contribution due to the local levels of the impurity. We have omitted the term F_{sw} in our calculations, because it leads only to some renormalization factor.[6] Then the magnetization curve can be determined from Eq. (2a) by

$$M=-\partial F/\partial H. \tag{3}$$

Finally, to make comparison with our experimental data we used $\chi=dM/dH$.

Figure 4 shows the calculated curves of the differential magnetic susceptibility of the $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ sample at different temperatures. It is evident from this figure that the model shows small ripples on the susceptibility peak at $T=0.6\,\mathrm{K}$ and only a further temperature decrease (up to 0.2 K)

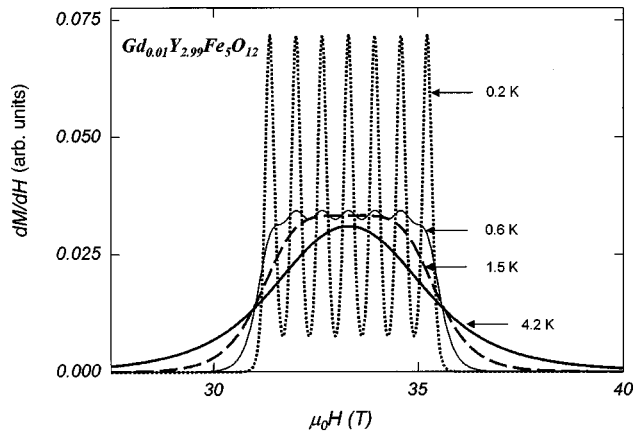

FIG. 4. The differential magnetic susceptibility versus field for $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ at different temperatures calculated in the framework of model (Ref. 6). The parameters used in the calculation are $\alpha=1.2\ \mathrm{cm}^{-1}$, $J_0=3.5\ \mathrm{cm}^{-1}$, $J=25\ \mathrm{cm}^{-1}$, and $z=7$.

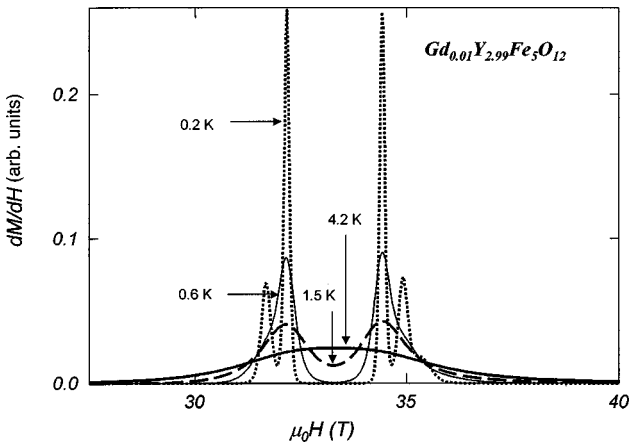

FIG. 5. The differential magnetic susceptibility versus field for $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ shown at different temperatures calculated within the framework of model (Ref. 7). The parameters used in the calculations are $\alpha=1.2\ \mathrm{cm}^{-1}$, $J_0=3.5\ \mathrm{cm}^{-1}$, $J=25\ \mathrm{cm}^{-1}$, $z=7$, $\kappa=0.59$, and $R=200$ Å.

leads to well pronounced oscillations. The total number of the susceptibility peaks is equal to $2^*S_{\mathrm{im}}=7$, in contradiction with the two peaks observed in our experiments. The temperature increase leads to broadening of the impurity levels and, as a result, to smoothing of the magnetization curve. Indeed, in order to get well pronounced oscillations of χ the temperature must be significantly less than α, which in fact determines the separation between two neighboring peaks in the susceptibility.[6]

The discrepancy between the simple model[6] (7 peaks) and experiments (2 peaks) is probably caused by ignoring the interaction between impurities in the simplest model. Increase of the impurity concentration must lead in the limit to impurity bands instead of levels.[7] In this transition to an impurity band, the levels will broaden which can prevent the appearance of the susceptibility oscillations. In what follows, a more sophisticated model will be used,[7] which considers an ideal gas of impurity *pairs* in a ferromagnetic matrix.

In the latter model, they took into account the interaction between impurities, which leads to the formation of impurity pairs and introduced the term $F_2(\mathbf{R})$ describing the free energy of the impurity pair. As a result, the term F_{im} [see Eq. (2b)] is replaced by the term F_{loc}, taking into account a Poisson distribution of the impurity pairs in a matrix

$$F_2(\mathbf{R})=-T\ln\sum_{m=0}^{4S_0}\sum_{l}\exp(-\beta\varepsilon_{ml}), \tag{4}$$

$$F_{\mathrm{loc}}=\frac{x}{2}\int F_2(\mathbf{R})W(\mathbf{R})d\mathbf{R},\quad W(\mathbf{R})=x\exp(-4\pi x\mathbf{R}^3/3). \tag{5}$$

here ε_{ml} are the energy levels of the bound states at the impurity pair, and $\mathbf{R}=\mathbf{r}_1-\mathbf{r}_2$ is the distance between two impurity ions (the lattice constant is assumed equal to unity), $W(\mathbf{R})$ is the probability of finding the second impurity at the distance R from the first one:

$$\varepsilon_{ml}=m\varepsilon_1+\frac{1}{2}l(l-1)\alpha+\frac{1}{2}(m-l)(m-l-1)\alpha\mp(l+1)\times\left(1-\frac{l}{2S_0}\right)\varepsilon_R\delta_{m,2l+1}, \tag{6}$$

where $\delta_{m,21+1}$ is the Kronecker symbol and the energy levels are assigned by the second index l in accordance with the classification given in Ref. 7. In other words, for given m ($0\leqslant m\leqslant 4S_0$ for a pair), some of the levels with the initial energy ε_m [see Eq. (2a)] break into a number of sublevels corresponding to a different distribution of magnons over the pair of impurities. The parameter l describes a number of magnons for one impurity ($0<l<2S_0$). The parameters ε_1 and ε_R are given by formulas

$$\varepsilon_1=g_0\mu_B H-zSJ_0-S_0\alpha, \tag{7}$$

$$\varepsilon_R=zS_0\frac{(J_0)^2}{J}\frac{n}{4\pi}\frac{e^{-\kappa R}}{R},\quad \kappa^2=n\frac{J_0}{J}. \tag{8}$$

Here n is equal to 2 and 3 for the bcc and fcc lattices, respectively. The radius of the bound state is $R_b\sim 1/\kappa$, and the average distance between impurities is $R_p\sim c^{-1/3}$. In addition, it is assumed in Ref. 7 that $c^{1/3}/\kappa\ll 1$, i.e., the radius of the bound state is small in comparison with the average distance between impurities. This condition is fulfilled for the garnet with $x=0.01$ ($R_b\approx 1.5$–1.9; $R_p\approx 6.7$). Figure 5 shows the results of the calculated field dependence of the differential magnetic susceptibility of the $Gd_{0.01}Y_{2.99}Fe_5O_{12}$ sample, taking into account the concentration broadening. The parameters used for this simulation are presented in the figure caption. It is evident that this model approaches the experiment much better; one initial peak of susceptibility at $T=4.2$ K splits into two peaks at $T=1.5$ K and $T=0.6$ K. Further temperature decrease causes additional splitting at $T=0.2$ K. One can conclude that interaction between impurities drastically changes the susceptibility pattern. Instead of 7

susceptibility peaks it is possible to observe two or four peaks depending on temperature. Nevertheless, the stepwise character of the magnetization curve remains and the differential magnetic susceptibility manifests quantum oscillations in fields near $\mu_0 H \sim 32$ T.

CONCLUSION

The appearance of field-induced oscillations in the magnetic susceptibility of a magnet with antiferromagnetic impurities is reported. This is a noncooperative quantum phenomenon, predicted a long time ago in Refs. 6 and 7. Numerical simulations, made in the framework of the sophisticated model[7] show a qualitative agreement with the experimental results, confirming this quantum phenomenon. The used models represent, to our mind, only the first approximation to a real picture of this phenomenon, and on the next stage a more realistic model based upon the results of this work should be developed, which could take into account clusters of impurities, the realistic magnon spectrum, etc.

This effect has a general character and may be observable under certain conditions (see Refs. 6,7) in a large number of ferro-or ferrimagnets with antiferromagnetic impurities.

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