Processes of spin-wave relaxation on paramagnetic impurities in antiferromagnets

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We investigate the following: a) nonlinear relaxation of spin waves (SW) in direct resonant transitions in a paramagnetic impurity, b) SW relaxation via two- and three-magnon processes with transitions in the impurity, c) nonlinear "slow" SW relaxation assuming the SW frequency to be small compared with the level spacing. In the study of the "slow" relaxation due to modulation of the spacing of the paramagnetic-impurity levels by the spin waves, it was observed that this modulation can cool the impurity subsystem. The calculation results are used to interpret the experimental data on parametric excitation of SW in antiferromagnetic crystals.

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§1. INTRODUCTION

The strong splitting of the levels of impurity paramagnetic ions in antiferromagnets, due to the lattice crystal field, to exchange interaction with the spins of the neighboring atoms, and to spin-orbit interaction, causes the spacing of the lower levels of paramagnetic ions to become comparable with the spin-wave (SW) frequencies. Because of this, besides the elastic scattering of the SW by the impurities, an important role is assumed in the SW relaxation by processes accompanied by energy transfer from the SW into the impurity subsystem. It is known that such processes make a substantial contribution to SW relaxation in ferromagnetic yttrium iron garnet (YIG) when the latter contains paramagnetic impurities (see Ref. 1). Crystals of the antiferromagnets used in experiments on parametric excitation of SW likewise contain usually a definite fraction of paramagnetic impurities,[2] and therefore a calculation of the linear and nonlinear relaxation of SW on paramagnetic impurites in antiferromagnets is of considerable interest.

One can single out several SW-relaxation mechanisms connected with energy transfer to the impurity subsystem. If the SW frequency coincides with the distance between two lower levels of the impurity, direct resonant transitions with absorption of one SW are allowed. There exist, however, also processes that do not require this equality. These include the two-magnon inelastic scattering of SW by an impurity, with a transition within the impurity ion, and also the three-magnon processes of coalescence and decay of SW, accompanied by a transition within the impurity. Finally, there exists also a mechanism of "slow" SW relaxation,[1] connected with modulation of the distance between the lower levels in the impurity paramagnetic ion by the spin wave.

The present paper is devoted to a systematic study of the contribution of such mechanisms to a linear and nonlinear SW relaxation. We emphasize that some of the problems that arises here were already discussed in the literature. The contribution made to linear SW relaxation by direct resonant transitions between impurity levels was calculated in Refs. 3–6. In Refs. 6 and 7 was constructed a phenomenological theory of linear "slow" relaxation on paramagnetic ions; this theory is valid when the distance between the lower levels in the impurity ion greatly exceeds the frequency of the modulating spin wave. In Ref. 8 is proposed a quantum theory of linear and nonlinear "slow" relaxations, which requires no definite relaxations between the SW frequency and the spacing of the lower level of the impurity, but which presupposes that the frequency of the relaxation of the population of the impurity levels $\gamma_{\parallel}$ is much less than the spin-wave frequency; this theory is therefore valid only at sufficiently low (helium) temperatures.

Taking the foregoing into account, we discuss in the present paper the following topics: a) linear relaxation of spin waves in direct resonant transitions in an impurity; b) relaxation of spin waves on account of two- and three-magnon processes at higher temperatures, assuming a low SW frequency compared with the interlevel distance. We note that the problem of calculating the nonlinear relaxation of spin waves is itself connected at the present time principally with the need for a theoretical interpretation of the numerous results obtained in experiments on parametric excitation of spin waves.

The energy spectrum of the parametric impurity ions in antiferromagnets is formed by the actions of the Coulomb interaction between the electrons within the atom, of the effective electric crystal field due to the particles surrounding the ion, of the external magnetic field, and of the exchange interaction with the spins of the neighboring atoms of the host lattice.

The action of the crystal field is always weaker than the Coulomb interaction of the ion with its electrons. The crystal field is regarded as "weak"[9] if it is incapable of breaking the bond between the orbital and spin moments of the entire unfilled electron shell. This case is realized, in particular, in rare-earth ions, since they have a deeply lying unfilled electron shell that determines the magnetic properties of the $4f$ ions and is screened by the outer $5s$ and $5p$ electrons. The crystal field is called "average"[9] if its action is stronger than the spin-orbit coupling but is much weaker than the interaction between the electrons inside the ion. A similar situation arises, e.g., in paramagnetic ions of the iron-group elements, since their unfilled electron shell is the outer one.

The character of the splitting of the energy levels of paramagnetic impurity ions by the crystal field is deter-

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mined to a considerable degree by the symmetry of the field. For impurity ions with an even number of electrons, the crystal field can lift completely the degeneracy of the level at a sufficiently low (e.g., rhombic) symmetry of this field. In the case of a half-integer spin the energy levels of the impurity paramagnetic ion remain at least doubly degenerate in the crystal field. This effect is the consequence of the general Kramers theory[9]: the electric forces are incapable of lifting completely the degeneracy of an energy level of a system containing an odd number of electrons. A magnetic field (including the exchange field), by lifting the degeneracy of the ground level of the field, can cause a splitting that lies in the microwave region.

Also possible are close non-Kramers doublets. If the symmetry of the crystal field is higher than rhombic, some of the levels produced under its influence remain doubly degenerate. Further small splitting of these levels is due to local distortions of the crystal field, caused by dislocations and other crystal-lattice defects. The resultant splittings have a random spread.

Another type of non-Kramers doublet consists of two singlet levels that turn out accidentally close to each other. Accidental approach of the levels is observed, e.g., in the energy spectrum of a Pr^{3+} ion in YIG.[1]

Calculation of the spectrum of the impurity paramagnetic ions is not the purpose of this paper, and we assume this spectrum to be given. The spectrum of the impurity ions Yb^{3+} in dodecahedral sites of the crystal lattice of the YIG were considered in Ref. 10, and for impurity terbium ions in YIG such a calculation was carried out in Ref. 11. The spectrum of the Fe^{2+} ion in an antiferromagnet of the "easy plane" type of rhombohedral symmetry, $MnCO_3$, was investigated in Ref. 12, and that of Mn^{2+} ions in the two-dimensional antiferromagnet K_2CoF_5 was investigated in Ref. 13.

In an experiment[14] on a sample of antiferromagnetic $MnCO_3$, containing according to Ref. 2 paramagnetic impurities with concentrations 0.35% Fe, 0.04% Ni and 0.01–0.45% Co, a sharp peak was observed in the spin-wave relaxation and can be attributed to direct resonant transitions between the levels in the impurity ions of one of the available types. It can be assumed that such impurity paramagnetic ions are the CO^{2+} ions, whose lowest level is separated from the remaining ones by several hundred reciprocal centimeters, turns out to be a Kramers doublet.[9] The Ni^{2+} and Fe^{2+} impurity ions have an even number of electrons in the unfilled shell and their energy levels are therefore fully split by the crystal field of the lattice of low rhombohedral symmetry; in this connection, excluding an unlikely random approach of the lower levels, one should expect the distance between the lower levels in such ions to exceed substantially the spin-wave frequency. Therefore direct transitions with absorption of spin waves by these ions are impossible, although they can take part in other processes of interaction with spin waves.

§2. SINGLE-MAGNON RELAXATION ON PARAMAGNETIC IMPURITIES

The expression for the contribution to the Hamiltonian that describes direct resonant transitions inside an impurity ion with absorption or emission of a spin wave is of the form

$$H_1 = \frac{1}{\mathcal{N}^{1/2}} \sum_{j\mathbf{k}} (\Psi_{j\mathbf{k}} b_\mathbf{k} a_j^+ + \text{H.c.}), \tag{1}$$

where $b_\mathbf{k}$ is the complex amplitude[1)] of a spin wave with wave vector, $\mathbf{k}$, and the operator a_j^+ effects the transition from the ground to the first excited state in an impurity located at the site $\mathbf{R}_j$ of the crystal lattice; $\mathcal{N}$ is the total number of sites in the crystal, and expressions for the amplitudes $\Psi_{j\mathbf{k}}$ are given in Ref. 8. We note that the operators a_j^+ and a_j, which pertain to the same site, have Fermi permutation relations, while the operators pertaining to different sites commute with one another.

The interaction of impurities with the thermostat (thermal phonons and magnons) will be taken into account phenomenologically, introducing the transverse $\gamma_\perp$ an the longitudinal $\gamma_\parallel$ relaxation frequencies in the corresponding kinetic equations for the population of the upper level of the impurity $n_j = \langle a_j{}^+ a_j \rangle$.

Starting from (1), we can obtain the following equations for the average spin-wave intensity $N_\mathbf{k} = \langle b_\mathbf{k}{}^* b_\mathbf{k} \rangle$, for population n_j, and for the quantity $\langle a_j \rangle$:

$$\dot{N}_\mathbf{k} = \frac{1}{\hbar \mathcal{N}^{1/2}} \sum_j \operatorname{Im}(\Psi_{j\mathbf{k}} \langle a_j^+ b_\mathbf{k} \rangle) + \ldots, \tag{2}$$

$$\dot{n}_j = -\gamma_\parallel (n_j - I_0) - \frac{1}{\hbar \mathcal{N}^{1/2}} \sum_\mathbf{k} \operatorname{Im}(\Psi_{j\mathbf{k}} \langle a_j^+ b_\mathbf{k} \rangle), \tag{3}$$

$$\frac{d}{dt} \langle \bar{a}_j \rangle = -i(\Omega_0 - \omega_\mathbf{k}) \langle \bar{a}_j \rangle - \gamma_\perp \langle \bar{a}_j \rangle - \frac{i}{\hbar \mathcal{N}^{1/2}} \sum_\mathbf{k} \Psi_{j\mathbf{k}} \langle \bar{b}_\mathbf{k} (1 - 2a_j^+ a_j) \rangle. \tag{4}$$

Here $I_0 = [\exp(\hbar\Omega_0/\Theta) + 1]^{-1}$ is the average equilibrium population of the upper level in a two-level atom with distance Ω_0 between the levels at a temperature Θ; $\omega_\mathbf{k}$ is the spinwave frequency. In Eq. (2) for $N_\mathbf{k}$ we did not write out explicitly the terms that describe the damping of the spin wave on account of other relaxation mechanisms, and the possible interaction with the electromagnetic parametric pumping. In Eq. (4) we have changed over to the "slow" quantities $\bar{a}_j$ and $\bar{b}_\mathbf{k}$ defined as

$$\bar{b}_\mathbf{k} = b_\mathbf{k} \exp(i\omega_\mathbf{k} t), \quad \bar{a}_j = a_j \exp(i\omega_\mathbf{k} t).$$

In the experiment, the characteristic spin-wave relaxation times are always larger by several orders of magnitude than the impurity relaxation time. It can therefore be assumed that the quantities n_j and $\langle \bar{a}_j \rangle$ attune themselves at each instant of time of the instantaneous value of the slow spin-wave amplitude $\bar{b}_\mathbf{k}$. By solving the obtained stationary equations for $\langle \bar{a}_j \rangle$ and n_j, assuming a low impurity density, we determine the steady-state population difference in the impurity ion $\sigma_j = 1 - 2n_j$,

$$\sigma = \sigma_0 \xi_c / (\xi_0 + \xi_c), \tag{5}$$

where $\sigma_0 = 1 - 2I_0$ is the equilibrium population density, and ξ_c is given by

$$\xi_c = \frac{\gamma_\parallel}{2\gamma_\perp}\left(\frac{\hbar}{|\Psi|}\right)^2\left[\gamma_\perp^2 + \left(\Omega_0 - \frac{\omega_p}{2}\right)^2\right]. \tag{6}$$

We assume that spin waves of frequency $\omega_{\mathbf{k}} = \omega_p/2$ have been parametrically excited in the crystal. The total number of the parametrically excited spin waves per lattice site is $\xi_0 = N_0/\mathcal{N}$. We note that owing to the fluctuations there exists a definite scatter $\delta\omega$ in the frequencies of the parametric spin waves; the size of this scatter, however, is always less than $\gamma_\perp$ and can be neglected.

According to (5), an increase in the number of parametric spin waves leads to a decrease in the population difference σ, i.e., to heating of the impurities.

The contribution to the spin-wave relaxation on account of direct resonant transitions is of the form

$$\Gamma_{\mathbf{k}} = c\left(\frac{|\Psi|}{\hbar}\right)^2 \sigma_0 \frac{\gamma_\perp}{\gamma_\perp^2 + (\Omega_0 - \omega_p/2)^2}\,\frac{\xi_c}{\xi_0 + \xi_c}, \tag{7}$$

where c is the dimensionless impurity density.

As seen from (7), with increasing number of parametric spin waves the relaxation frequency $\Gamma_{\mathbf{k}}$ decreases, i.e., the nonlinear damping obtained by us turns out to be negative in a differential sense, with $\Gamma_{\mathbf{k}} \to 0$ at $\xi_0 \gg \xi_c$.

When the spin-wave frequency $\omega_{\mathbf{k}} = \omega_p/2$ exactly coincides with the distance Ω_0 between the lower levels of the impurity, the spin-wave density reaches its maximum value. If the distance Ω_0 between the impurity levels depends on the external magnetic field we can, by varying this field, make Ω_0 equal to $\omega_p/2$, and the plot of $\Gamma_{\mathbf{k}}$ against the magnetic field should show a sharp peak with a characteristic frequency width of the order of $\gamma_\perp$.

In experiments on parametric excitation of spin waves in antiferromagnets,[14] there is actually observed a clearly pronounced maximum on the dependence of the turned-off part of the spin-wave relaxation on the external magnetic field, but its width greatly exceeds the frequency the transverse relaxation for the impurities. We assume that this is the consequence of the inhomogeneous broadening of the resonant absorption line, due to the random scatter of the distance between the lower levels in the impurity ions. The impurity ion that interacts strongly with the lattice and with the magnetic moments of the neighboring atoms is always sensitive to small surrounding distortions that can be produced by dislocations, vacancies, and other defects. As a result, the resonant frequency of each ion shifts slightly relative to its value for the ideal lattice.

To take into account the differences in the frequency of the transitions between the levels of the impurity ions, we shall assume that the frequencies of the impurities have a scatter about the fundamental frequency Ω_0, and that the distance between the lower levels of the impurity at the site $\mathbf{R}_j$ can be represented in the form $\Omega_j = \Omega_0 + \varepsilon_j$, where ε_j is a random quantity with a certain distribution $P = P(\varepsilon)$. We assume for the sake of argument that this distribution is Lorentzian

$$P(\varepsilon) = \bar{\varepsilon}/(\bar{\varepsilon}^2 + \varepsilon^2). \tag{8}$$

Repeating the reasoning used to derive (7), we obtain after additional averaging over the frequency scatter the following general expression for the nonlinear relaxation of spin waves in the case of inhomogeneous broadening:

$$\begin{aligned}\Gamma_{\mathbf{k}} = \frac{c(\Psi/\hbar)^2\sigma_0\pi\varepsilon\gamma_\perp}{(\gamma_\perp^2+\delta^2-\varepsilon^2)^2+4\delta^2\varepsilon^2}\Bigg\{&\left(\frac{\gamma_\perp^2+\delta^2-\varepsilon^2}{\varepsilon} - \frac{\gamma_\perp^2-\varepsilon^2-\delta^2}{\gamma_\perp}\right)\\ &- \frac{4\xi_0(\Psi/\hbar)^2}{\gamma_\parallel}\Bigg\{-\frac{1}{\gamma_\perp^2}(\gamma_\perp^2-\varepsilon^2-2\delta^2)\\ &+ \frac{\gamma_\perp}{(\gamma_\perp^2+\delta^2-\varepsilon^2)^2+4\delta^2\varepsilon^2}\Bigg[-4\delta^2\varepsilon + \frac{(\gamma_\perp^2+\delta^2-\varepsilon^2)^2}{\varepsilon}\\ &- \frac{1}{\gamma_\perp}[(\gamma_\perp^2+\delta^2-\varepsilon^2)(\gamma_\perp^2-\varepsilon^2-3\delta^2) - 4\delta^2\varepsilon^2]\Bigg]\Bigg\}\Bigg\}, \end{aligned} \tag{9}$$

where it is assumed that the excited spin waves have a frequency $\omega_{\mathbf{k}} = \omega_p/2$, and we have introduced the frequency detuning $\delta = \Omega_0 - \omega_p/2$.

In the limiting case when the frequency scatter is extremely small ($\bar{\varepsilon} \ll \gamma_\perp$), Eq. (9) goes over into expression (7) above. In the opposite limiting case $\varepsilon \gg \gamma_\perp$, expression (9) can be represented in the simpler form

$$\Gamma_{\mathbf{k}} = c\pi\left(\frac{\Psi}{\hbar}\right)^2 \sigma_0 \frac{\varepsilon}{\varepsilon^2+\delta^2}\left(1 - 4\left(\frac{\Psi}{\hbar}\right)^2 \xi_0 \frac{\varepsilon^2+2\delta^2}{\gamma_\perp\gamma_\parallel}\right). \tag{10}$$

Consequently the spin-wave damping $\Gamma_{\mathbf{k}}$ duplicates in this limiting case the Lorentzian frequency distribution (8), and the width of the resonant peak is equal to the average impurity-frequency scatter.

We note that in (9) and (10) we expanded in terms of the parameter ξ_0/ξ_c. These equations are therefore valid for relatively small numbers of parametrically excited spin waves (PESW) ξ_0. In the opposite limiting case when $\xi_0 \gg \xi_c$, the relaxation is completely turned off on account of direct transitions [cf. Eq. (7)].

§3. "SLOW" RELAXATION OF SPIN WAVES

In this section we discuss the contribution made to spin-wave relaxation by their modulation of the distance between the levels in the impurity ions. In contrast to Ref. 8, we do not assume here that the longitudinal relaxation of the impurities $\gamma_\parallel$ is small compared with the spin-wave frequency $\omega_{\mathbf{k}}$, and therefore the results of the calculations are applicable also to the case of higher temperatures. At the same time, we assume that the modulation is smooth, i.e., $\omega_{\mathbf{k}} \ll \Omega_0$; this assumption was not significant in Ref. 8.

The Hamiltonian that describes the modulation, by the spin waves, of the distance between the levels in the impurity ions, is of the form

$$H = \sum_j \hbar(\Omega_0 + \delta\Omega)a_j^+ a_j, \tag{11}$$

where

$$\delta\Omega = \frac{1}{\hbar\mathcal{N}^{1/2}}\sum_{j\mathbf{k}}(\Phi_{j\mathbf{k}}b_{\mathbf{k}} + \Phi_{j\mathbf{k}}^* b_{\mathbf{k}}^*). \tag{12}$$

Expressions for the amplitude of the interaction $\Phi_{j\mathbf{k}}$ are given in Ref. 8.

The system of impurity ions interacts also with the thermostat (the thermal phonons and magnons). This interaction ensures relaxation of the population of the upper levels of the impurity ions to the equilibrium thermal value

$$I=[\exp(\hbar\Omega/\Theta)+1]^{-1} \tag{13}$$

at a distance Ω between the levels in the ions. The population relaxation is described by the kinetic equation

$$dn/dt=-\gamma_{\parallel}(n-I), \tag{14}$$

where $\gamma_{\parallel}$ is the longitudinal-relaxation frequency. Expressions for $\gamma_{\parallel}$ were obtained by us in Ref. 8. We note that $\gamma_{\parallel} = \gamma_{\parallel}(\Omega)$, i.e., it is a certain function of the distance between the impurity levels.

Modulation of the distance between the levels by spin waves means that the quantity $\Omega = \Omega_0 + \delta\Omega(t)$ oscillates with time. If, as we assume, the characteristic modulation frequencies $\omega_{\mathbf{k}}$ are low compared with Ω_0, such a smooth change of Ω can be taken into account within the framework of the kinetic equation (14), namely, it can be assumed that at each instant of time the population n relaxes to the equilibrium value $I[\Omega_0 + \delta\Omega(t)]$, corresponding to the distance between the levels at the given instant of time, and this takes place at a rate $\gamma_{\parallel}[\Omega_0 + \delta\Omega(t)]$.

We shall assume that the intensity of excitation of the spin waves is not too high, so that the following condition is satisfied[2)]

$$\hbar|\delta\Omega|/\Theta\ll 1. \tag{15}$$

Then the quantities $I(\Omega_0 + \delta\Omega)$ and $\gamma_{\parallel}(\Omega_0 + \delta\Omega)$ can be expanded in powers of $\delta\Omega$, i.e., actually in powers of the amplitudes $b_{\mathbf{k}}$ and $b_{\mathbf{k}}^*$ of the spin waves:

$$\begin{aligned} I(\Omega_0+\delta\Omega)&=I_0+I_0'\delta\Omega+{}^1/_2I_0''\delta\Omega^2+{}^1/_6I_0'''\delta\Omega^3+\ldots,\\ \gamma_{\parallel}(\Omega_0+\delta\Omega)&=\gamma_{\parallel}(\Omega_0)+\gamma_{\parallel}'\delta\Omega+{}^1/_2\gamma_{\parallel}''\delta\Omega^2+{}^1/_6\gamma_{\parallel}'''\delta\Omega^3+\ldots. \end{aligned} \tag{16}$$

Substituting these expansions in (14) we can seek the solution of the obtained equation also in the form of a series in the short-wave amplitudes. Equating the terms in the left and right sides of this equation, which are of the same order in the spin-wave amplitude and which oscillate in time in accordance with the same law, we find the system of equations that connects these corrections.

The equation for finding the first-order correction $\delta n_1(t)$ to the equilibrium value of the population $I_0 = I(\Omega_0)$ is of the form

$$d\delta n_1/dt=-\gamma_{\parallel}(\Omega_0)\delta n_1+\gamma_{\parallel}(\Omega_0)I'\delta\Omega(t).$$

The solution of this equation is given by

$$\delta n_1=\frac{\gamma_{\parallel}(\Omega_0)I'}{\hbar\mathcal{N}^{1/2}}\sum_{\mathbf{k}}\left\{\frac{\Phi_{j\mathbf{k}}b_{\mathbf{k}}}{\gamma_{\parallel}(\Omega_0)-i\omega_{\mathbf{k}}}+\text{c.c.}\right\}. \tag{17}$$

The average spin-wave intensity $N_{\mathbf{k}} = \langle b_{\mathbf{k}}^* b_{\mathbf{k}}\rangle$ varies with time as a result of the interaction (11) like

$$\dot{N}_{\mathbf{k}}=-(2/\hbar\mathcal{N}^{1/2})\sum_j\operatorname{Im}(\Phi_{j\mathbf{k}}\langle b_{\mathbf{k}}n_j\rangle). \tag{18}$$

If we substitute in (18) $n_j = I_0 + \delta n_1$ and recognize that $\langle b_{\mathbf{k}}\rangle = \langle b_{\mathbf{k}}^*\rangle = 0$, we obtain

$$\dot{N}_{\mathbf{k}}=-\Gamma_{\mathbf{k}}^0N_{\mathbf{k}}, \tag{19}$$

where the linear relaxation of the spin waves on account of the modulation of the interlevel distance is of the form

$$\Gamma_{\mathbf{k}}^0=-2c\left(\frac{|\Phi|}{\hbar}\right)^2\gamma_{\parallel}(\Omega_0)I'\frac{\omega_{\mathbf{k}}}{\gamma_{\parallel}^2(\Omega_0)+\omega_{\mathbf{k}}^2}. \tag{20}$$

Here c is the dimensionless impurity density (the ratio of the number of impurity ions to the total number of sites in the crystal):

$$I'=\frac{dI}{d\Omega}=-\frac{\hbar}{\Theta}\exp\left(\frac{\hbar\Omega_0}{\Theta}\right)I^2(\Omega_0). \tag{21}$$

The expression obtained for the linear relaxation agrees with the result of the phenomenological "slow"-relaxation theory[6] constructed for ferromagnets.

The temperature dependence of the linear relaxation $\Gamma_{\mathbf{k}}^0$ is determined by the temperature dependence of the factor I' and by the longitudinal impurity relaxation frequency $\gamma_{\parallel}$. The last factor in (20) goes through a maximum at a temperature Θ satisfying the condition $\gamma_{\parallel}(\Omega_0, \Theta) = \omega_{\mathbf{k}}$. Taking into account, however, the temperature dependence of the other factors, we cannot state that the relaxation $\Gamma_{\mathbf{k}}^0$ will reach a maximum precisely at this temperature. Bearing this in mind, we have used Eq. (20) for a numerical calculation of $\Gamma_{\mathbf{k}}^0$ at the following parameters corresponding to the antiferromagnet $FeBO_3$; $c = 0.01\%$, $\omega_{\mathbf{k}} = \omega_p/2 = 10^{11}$ sec^{-1}, $\Phi = 10^{-14}$ erg, $\omega_{\mathbf{k}}^2 = \omega_0^2 + \omega_E^2(ak)^2$, $\omega_E = 10^{13}$ sec^{-1}, $a = 10^{-8}$ cm, $\omega_0^2 = g^2[H(H + H_D)]$, $H_D = 100$ kOe, and $H = 0.3$ kOe.

We have assumed that the longitudinal relaxation $\gamma_{\parallel}$ is determined by the direct transition between the levels, with emission of spin waves or phonons, and used the expressions given in Ref. 8 for $\gamma_{\parallel}$. In the calculation of $\gamma_{\parallel}(\Omega)$ we have assumed $\Psi^{Ph} = 10^{-7}$ erg/cm and $\Psi^m = 10^{-15}$ erg; the crystal density was $\rho = 5$ g/cm^3, the phonon velocity $v = 10^5$ cm/sec, and the Debye temperature $\Theta_D = 10^{-14}$ erg. The calculated temperature dependence of $\Gamma_{\mathbf{k}}^0$ at various distances between the levels Ω_0 are shown in Fig. 1.

The nonlinear effects are determined by corrections of

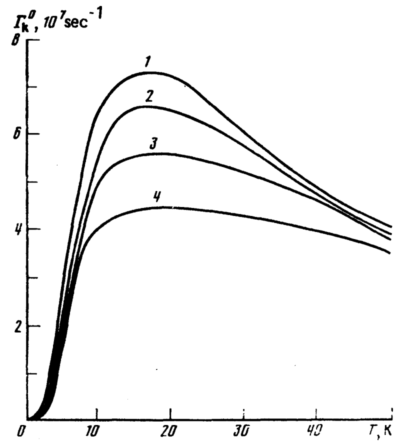

FIG. 1. Dependence of the linear frequency of the "slow" spin-wave relaxation Γ_k^0 on the temperature for antiferromagnetic $FeBO_3$. Calculation by formula (20): 1 — $\Omega_0 = 16\cdot10^{11}$ sec^{-1}, 2 — $\Omega_0 = 14\times10^{11}$ sec^{-1}, 3 — $\Omega_0 = 12\cdot10^{11}$, 4 — $\Omega_0 = 10\cdot10^{11}$ sec^{-1}.

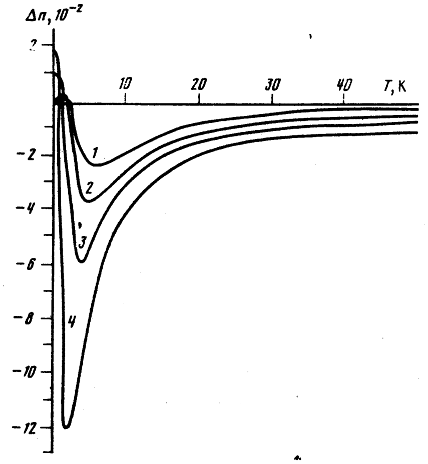

FIG. 2. Dependence of the deviation Δn of the population of the upper impurity level on the temperature. Calculation by formula (22): 1 — $\Omega_0 = 16\cdot 10^{11}$ sec^{-1}, 2 — $\Omega_0 = 8\cdot 10^{11}$ sec^{-1}, 3 — $\Omega_0 = 6\cdot 10^{11}$ sec^{-1}, 4 — $\Omega_0 = 4\cdot 10^{11}$ sec^{-1}.

higher order to the equilibrium population of the upper level. In particular, the change of the population averaged over the spin-wave period is given by the time-independent second-order correction

$$\Delta n \approx \sum_{\mathbf{k}} b_{\mathbf{k}}^* b_{\mathbf{k}}.$$

A consistent calculation leads to the following expression for Δn:

$$\Delta n = 2\left(\frac{|\Phi|}{\hbar}\right)^2 \xi_0 \left\{\frac{1}{2} I'' + \frac{\gamma_\parallel'}{\gamma_\parallel} I' \frac{\omega_{\mathbf{k}}^2}{\gamma_\parallel^2+\omega_{\mathbf{k}}^2}\right\}. \tag{22}$$

At low temperatures, when $\gamma_\parallel < \omega_{\mathbf{k}}$, this expression goes over into the corresponding formula of Ref. 8 if it is assumed in the latter that $\Omega_0 \gg \omega_{\mathbf{k}}$. Plots of Δn against the temperature Θ at different frequencies of the transition between the levels are shown in Fig. 2. It can be seen that the action of the intense spin waves can lead both to heating ($\Delta n > 0$) and to cooling ($\Delta n < 0$) of the impurity subsystem. We emphasize that effects of cooling of different two-level systems in interactions with intense waves were discussed also in Refs. 8, 15, and 16.

To calculate the nonlinear damping it is necessary to calculate the third-order correction

$$\delta n_3 \sim \left(\sum_{\mathbf{k}'} b_{\mathbf{k}'}^* b_{\mathbf{k}'}\right) b_{\mathbf{k}} \tag{23}$$

to the equilibrium population of the impurity level. Calculation shows that for a nonlinear damping coefficient η, defined by the relation $\Gamma_{\mathbf{k}} = \Gamma_{\mathbf{k}}^0 + \eta\xi_0$, the following expression holds

$$\eta = c\left(\frac{|\Phi|}{\hbar}\right)^4 \omega_{\mathbf{k}} \left\{ 4\gamma_\parallel'' I' \frac{\gamma_\parallel^2}{(\gamma_\parallel^2-\omega_{\mathbf{k}}^2)^2 + 4\omega_{\mathbf{k}}^2\gamma_\parallel^2} - \frac{1}{\gamma_\parallel^2+\omega_{\mathbf{k}}^2}\left[\gamma_\parallel I''' + 2\gamma_\parallel'' I' - 4\frac{(\gamma_\parallel')^2 I'}{\gamma_\parallel}\frac{\omega_{\mathbf{k}}^2}{\gamma_\parallel^2+\omega_{\mathbf{k}}^2}\right]\right\}. \tag{24}$$

In the derivation of (24) we have assumed that all the excited spin waves have a frequency $\omega_{\mathbf{k}}$; their number per lattice site is $\xi_0 = N_0/\mathcal{N}$.

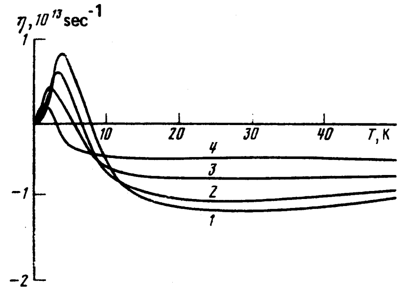

FIG. 3. Dependence of the coefficient of nonlinear damping η on the temperature. Calculation by formula (24) 1 — $\Omega_0 = 10\cdot 10^{11}$ sec^{-1}, 2 — $\Omega_0 = 8\cdot 10^{11}$ sec^{-1}, 3 — $\Omega_0 = 6\cdot 10^{11}$ sec^{-1}, 4 — $\Omega_0 = 4\cdot 10^{11}$ sec^{-1}.

The dependence of the coefficient of nonlinear damping on the temperature at different values of the distance between the levels in impurity ions is shown in Fig. 3. At sufficiently high temperatures the coefficient η is always negative and tends to zero with increasing temperature. At low temperatures, the nonlinear damping reverses sign and becomes positive.

We recall that Eq. (24) was obtained under the assumption that the modulation frequency is much lower than the distance between impurity levels, i.e., $\omega_{\mathbf{k}} \ll \Omega_0$. For comparison we present also an expression for the nonlinear damping coefficient at relatively low temperatures (when $\gamma_\parallel \ll \omega_{\mathbf{k}}$), which is valid at all relations between[3)] $\omega_{\mathbf{k}}$ and Ω_0:

$$\begin{aligned}\eta = c\left(\frac{2\Phi}{\hbar\omega_p}\right)^4 \Bigg\{ & \gamma_\parallel^{-1}(\Omega_0)\left[\gamma_\parallel\left(\Omega_0+\frac{\omega_p}{2}\right) - \gamma_\parallel\left(\left|\Omega_0-\frac{\omega_p}{2}\right|\right)\right] \\ & \times\left[\gamma_\parallel\left(\left|\Omega_0-\frac{\omega_p}{2}\right|\right)\left(I\left(\Omega_0-\frac{\omega_p}{2}\right)-I(\Omega_0)\right)\right. \\ & \left. -\gamma_\parallel\left(\Omega_0+\frac{\omega_p}{2}\right)\left(I(\Omega_0)-I\left(\Omega_0+\frac{\omega_p}{2}\right)\right)\right] \\ & +\frac{1}{3}\Bigg[\gamma_\parallel(\Omega_0+\omega_p)\left(-I(\Omega_0+\omega_p)+I(\Omega_0)\right) - \gamma_\parallel(|\Omega_0-\omega_p|)\left(I(\Omega_0)\right. \\ & \left. -I(\Omega_0-\omega_p)\right) - 2\left[\gamma_\parallel\left(\Omega_0+\frac{\omega_p}{2}\right)\left(I(\Omega_0)-I\left(\Omega_0+\frac{\omega_p}{2}\right)\right)\right. \\ & \left.\left. -\gamma_\parallel\left(\left|\Omega_0-\frac{\omega_p}{2}\right|\right)\left(I(\Omega_0)-I\left(\Omega_0-\frac{\omega_p}{2}\right)\right)\right]\Bigg]\Bigg\}.\end{aligned} \tag{25}$$

It is easy to verify that at $\omega_{\mathbf{k}} \ll \Omega_0$ it goes over into (24).

§4. TWO- AND THREE-MAGNON RELAXATION OF SPIN WAVES ON PARAMAGNETIC IMPURITIES

The Hamiltonian that describes two-magnon Raman scattering of spin waves by paramagnetic impurities, at which a transition takes place also between levels in the impurity ion, is of the form

$$H_2 = \frac{1}{\mathcal{N}}\sum_{j\mathbf{k}_1\mathbf{k}_2}\{B_{j\mathbf{k}_1\mathbf{k}_2} b_{\mathbf{k}_1}^* b_{\mathbf{k}_2} a_j^+ + \text{H.c.}\}. \tag{26}$$

The interaction amplitude $B_{j\mathbf{k}_1\mathbf{k}_2}$ for an easy-plane antiferromagnet is given by the expression

$B_{jk_1k_2}$

$$=\frac{1}{8}\frac{\omega_E}{\omega_k}(\sin\theta_1\sin\varphi_1\Lambda_{zz}^{(1)}-\sin\theta_2\sin\varphi_2\Lambda_{zz}^{(2)}+i\sin\theta_1\cos\varphi_1\Lambda_{zz}^{\prime(1)}$$

$$-i\sin\theta_2\cos\varphi_2\Lambda_{zz}^{\prime(2)})\exp\{i(\mathbf{k}_1-\mathbf{k}_2)\mathbf{R}_j\}, \qquad (27)$$

where (θ_1, φ_1) and (θ_2, φ_2) are the Euler angles of the magnetization of the two sublattices of the antiferromagnet in a coordinate system whose z axis is directed along the magnetic moment of the impurities, and $\hat{\Lambda}$ (1, 2) are the exchange-interaction tensors (see the Appendix in Ref. 8).

The Hamiltonian (26) corresponds to the following kinetic equations for the population n_j of the upper level of the impurity and for the intensity $N_\mathbf{k}$ of the spin waves with spin vector $\mathbf{k}$:

$$\dot{n}_j=\mathscr{L}_j\{n_j, N_\mathbf{k}\}-\gamma_\parallel(n_j-I_0), \quad \dot{N}_\mathbf{k}=\mathscr{L}_\mathbf{k}\{n_j, N_\mathbf{k}\}-\Gamma_\mathbf{k}(N_\mathbf{k}-\bar{N}_\mathbf{k}), \qquad (28)$$

where the collision integrals $\mathscr{L}_j$ and $\mathscr{L}_\mathbf{k}$ are of the form

$$\mathscr{L}_j=\frac{2\pi}{\hbar^2\mathscr{N}^2}\sum_{\mathbf{k}_1\mathbf{k}_2}|B_{j\mathbf{k}_1\mathbf{k}_2}|^2\{(N_{\mathbf{k}_1}+1)N_{\mathbf{k}_2}(1-n_j)$$

$$-N_{\mathbf{k}_1}(N_{\mathbf{k}_2}+1)n_j\}\delta(\omega_{\mathbf{k}_1}-\omega_{\mathbf{k}_2}+\Omega_0), \qquad (29)$$

$$\mathscr{L}_\mathbf{k}=\frac{2\pi}{\hbar^2\mathscr{N}^2}\sum_{j\mathbf{k}_1}|B_{j\mathbf{k}\mathbf{k}_1}|^2\{(N_\mathbf{k}+1)N_{\mathbf{k}_1}(1-n_j)$$

$$-N_\mathbf{k}(N_{\mathbf{k}_1}+1)n_j\}\delta(\omega_\mathbf{k}-\omega_{\mathbf{k}_1}+\Omega_0)$$

$$+\frac{2\pi}{\hbar^2\mathscr{N}^2}\sum_{j\mathbf{k}_1}|B_{j\mathbf{k}\mathbf{k}_1}|^2\{(N_\mathbf{k}+1)N_{\mathbf{k}_1}n_j$$

$$-N_\mathbf{k}(N_{\mathbf{k}_1}+1)(1-n_j)\}\delta(\omega_\mathbf{k}-\omega_{\mathbf{k}_1}-\Omega_0). \qquad (30)$$

We note that a wave with a given wave vector $\mathbf{k}$ can participate in two types of processes. It can vanish with production of another spin excited state (the secondary spin waves have in this case a frequency $\omega_{\mathbf{k}_1}=(\omega_\mathbf{k}-\Omega_0)$. Also possible is vanishing of the given wave with production of a secondary wave and transition of the impurity from the excited state into the ground state (then $\omega_{\mathbf{k}_1}=\omega_\mathbf{k}+\Omega_0$).

We have also taken into account in the kinetic equations (28) the linear relaxation of the spin wave and of the impurities, due to the interaction with the thermostat.

The parametrically excited spin waves fill a narrow resonant layer of width $\varkappa$ near a sphere of radius $k_0(\omega_{\mathbf{k}_0}=\omega_p/2)$ in $\mathbf{k}$-space. The intense spin waves should alter in this case the spin-wave density in two regions: 1) in a narrow layer of $\mathbf{k}$-space near the sphere corresponding to the frequency $\omega_\mathbf{k}=\omega_p/2-\Omega_0$, and 2) in a similar layer near a sphere on which $\omega_\mathbf{k}=\omega_p/2+\Omega_0$. We denote the total number of the spin waves in the first and second regions by N_1 and N_2 and introduce the quantities $\xi_{1,2}=N_{1,2}/\mathscr{N}$. If we take into account in (28) only processes in which PESW takes part, we obtain the following equations for the average population n of the upper level of the impurities and for the values of ξ_1 and ξ_2:

$$\dot{n}=\xi_0[R_1\xi_1\sigma+R_2\xi_2\sigma+p_1R_1(1-n)-p_2R_2n]-\gamma_\parallel(n-I_0),$$
$$\dot{\xi}_1=cR_1\xi_0[\xi_1\sigma+p_1(1-n)]-\Gamma_s(\xi_1-\xi_1^0), \qquad (31)$$
$$\dot{\xi}_2=-cR_2\xi_0[\xi_2\sigma-p_2n]-\Gamma_s(\xi_2-\xi_2^0).$$

Here $\sigma=1-2n$ is the difference between the populations of the lower and upper levels of the impurities, and the coefficients R and p take the form

$$R_{1,2}=2\pi|B|^2/\hbar^2\varkappa v_{1,2}, \quad p_{1,2}=(1/2\pi)\varkappa k_{1,2}a^2, \qquad (32)$$

where $\mathbf{k}_{1,2}$ is the wave vector of a spin wave with frequency $\omega_{1,2}=\omega_p/2\mp\Omega_0$, and $v_{1,2}$ is the velocity of this spin wave.

We are interested in stationary solutions of Eqs. (31), which are established at a given number ξ_0 of parameterically excited waves. The behavior of the quantities close to $\omega_1=\omega_p/2-\Omega_0$ and $\omega_2=\omega_p/2+\Omega_0$, as functions of the change of ξ_0, turns out to be quite different. To demonstrate this, it is convenient to solve the last two equations of (31) for ξ_1 and ξ_2, assuming that the population difference σ is known:

$$\xi_1=\xi_1^0\frac{1+(p_1/2\xi_1^0)(cR_1/\Gamma_s)(1+\sigma)\xi_0}{1-(cR_1/\Gamma_s)\sigma\xi_0}, \qquad (33)$$

$$\xi_2=\xi_2^0\frac{1+(p_2/2\xi_2^0)(cR_2/\Gamma_s)(1-\sigma)\xi_0}{1+(cR_2/\Gamma_s)\sigma\xi_0}. \qquad (34)$$

Numerical estimates show readily that the coefficients of ξ_0 in the denominators of (33) an (34) greatly exceed the coefficients of ξ_0 in the numerators of these expressions. Thus, the behavior of the quantities ξ_1 and ξ_2 with increasing ξ_0 is determined mainly by the changes in the denominators of (33) and (34). Consequently, with increasing number ξ_0 of the PESW, the number of spin waves with frequencies $\omega_\mathbf{k}=\omega_p/2+\Omega_0$ decreases in comparison with the thermal equilibrium level, whereas the number of secondary spin waves with frequencies $\omega_\mathbf{k}=\omega_p/2-\Omega_0$ increases, and at values of σ close to $(\Gamma_s/cR_1\xi_0)$, the quantity ξ_1 exceeds considerably the thermal level ξ_1^0.

The contribution made to the relaxation frequency of the PESW by the direct and inverse processes of the two types is given by the expression

$$\Delta\Gamma_k=c[R_1\xi_1-R_2\xi_2)\sigma-{}^1\!/_2R_1p_1(1+\sigma)+{}^1\!/_2R_2p_2(1-\sigma)]. \qquad (35)$$

Taking into account the remarks made in the calculation of the nonlinear phenomena at large numbers of PESW, we neglect in (35) the terms containing ξ_2.

If we neglect the terms containing ξ_2 in (31), the stationary population difference σ of the impurity levels is determined by the quadratic equation

$$^1\!/_2\gamma_\parallel(\sigma_0-\sigma)[c(R_1/\Gamma_s)\xi_0\sigma-1]={}^1\!/_2p_1(1-\sigma)R_1\xi_0. \qquad (36)$$

The quantity p_1 in the right-hand side of this equation is extremely small, $p_1\sim(\varkappa/k_1)k_1^2a^2$; it is important to take it into account only for a correct choice of one of the two roots of Eq. (36) on passing through the value $\xi_0=\xi_c$,

$$\xi_c=\Gamma_s/cR_1\sigma_0, \qquad (37)$$

when a crossing of the roots of Eq. (36) with a zero right hand side would take place. Choosing that root of Eq. (36) which vanishes at σ_0 in the absence of PESW (i.e., at $\xi_0 = 0$), we obtain[4)]

$$\sigma=\sigma_0, \quad \xi_0<\xi_c; \qquad \sigma=\sigma_0(\xi_c/\xi_0), \quad \xi_0\geqslant\xi_c. \tag{38}$$

The total number of secondary spin waves concentrated near the resonance surface $\omega_{\mathbf{k}} = \omega_p/2 - \Omega_0$ in $\mathbf{k}$-space is given in this same approximation by the expression

$$\xi_1=0, \quad \xi_0<\xi_c; \qquad \xi_1=\frac{c\gamma_{\parallel}}{2\Gamma_s}\sigma_0\left(1-\frac{\xi_c}{\xi_0}\right), \quad \xi_0\geqslant\xi_c. \tag{39}$$

According to (38), at high pump levels, when the number ξ_0 of the PESW exceeds the critical value ξ_c, the impurity system is heated and saturates ($\sigma = 0$) in the limit $\xi_0 \gg \xi_c$. Simultaneously, the number of secondary spin waves near the resonance surface $\omega_{\mathbf{k}} = \omega_p/2 - \Omega_0$ increases. It tends as $\xi_0 \to \infty$ to the limit

$$\xi_1^{max}=c\gamma_{\parallel}\sigma_0/2\Gamma_s.$$

We recall also that the spin-wave density near the surface $\omega_{\mathbf{k}} = \omega_p/2 + \Omega_0$ decreases as $\xi_0 \to \infty$ from the thermal level to zero [see (34)]. Substituting in (35) the expressions obtained for the difference of the populations (38) and for the number of secondary SW (39), we arrive at the following expressions for the correction to the PESW damping[5)]:

$$\Delta\Gamma_k=0, \quad \xi_0<\xi_c; \qquad \Delta\Gamma_k=\frac{c\gamma_{\parallel}\sigma_0}{2\xi_0}\left(1-\frac{\xi_c}{\xi_0}\right), \quad \xi_0>\xi_c. \tag{40}$$

The contribution $\Delta\Gamma_k$ reaches a maximum value

$$\Delta\Gamma_k^{max}=\frac{1}{4}c\frac{\sigma_0}{\xi_c}\gamma_{\parallel} \tag{41}$$

at $\xi_0 = 2\xi_c$.

Thus, if the crystal contains parametric rapidly relaxing impurities, they can play the role of an intermediate channel through which the PESW dissipate their energy in the thermostat. We note that effective inclusion of the relaxation $\Delta\Gamma_k$ due to two-magnon processes as well as heating of the impurity subsystem are subject to a threshold number of PESW. The point is that the decay of a PESW into a secondary spin wave with simultaneous excitation of the impurity begins to "operate" effectively when the rate of transitions of the impurities from the ground state to the excited one, due to such a proceses, begins to exceed the excited-to-ground-state transition rate that ensures energy dissipation of the impurities in the thermostat. After the PESW reach a number ξ_0 corresponding to the critical value ξ_c, the impurity system begins to become strongly heated. The heating continues until a temperature of the impurity subsystem is established such that the critical value ξ_c becomes comparable with the specified value ξ_0.

Numerical estimates for the threshold number of PESW in a two-magnon process at typical values of the parameters $c = 0.01\%$, $\Gamma_s = 10^5$ sec^{-1}, $\omega_p = 10^{11}$ sec^{-1}, $|B| = 10^{-15}$ erg, and $a = 10^{-8}$ cm yield the following result:

$$\xi_c=\left(\frac{\hbar}{|B|}\right)^2\frac{\Gamma_s\varkappa v_A}{\pi c\sigma_0}\sim 10^{-7}-10^{-8}, \tag{42}$$

which corresponds to a perfectly feasible, in experiments on parallel pumping, excess above the threshold of parametric excitation.

As already noted in §2, one should expect in a real crystal a certain scatter of the distances between the levels in different impurities. We have estimated the role of this scatter for the considered two-magnon process.

Assume that the distance between the lower levels of the impurity ion located at the site $\mathbf{R}_j$ is $\Omega_j = \Omega_0 + \varepsilon_j$, where ε_j is a random quantity with a distribution

$$P(\varepsilon)=0, \quad |\varepsilon|>\bar{\varepsilon}; \qquad P(\varepsilon)=1/2\bar{\varepsilon}, \quad |\varepsilon|<\bar{\varepsilon}. \tag{43}$$

Calculating the contribution of ε_k to the PESW relaxation we find, after an additional averaging over the frequency scatter, that the expression for $\Delta\Gamma_k$ is given as before by formula (40) in this case, provided, however, that the quantity σ_0 in (40) and (47) is replaced by the equilibrium impurity-population difference averaged over the frequency scatter:

$$\langle\sigma_0\rangle=\frac{\Theta}{\hbar\bar{\varepsilon}}\ln\left[\frac{\operatorname{ch}(\hbar(\Omega+\bar{\varepsilon})/2\Theta)}{\operatorname{ch}(\hbar(\Omega-\bar{\varepsilon})/2\Theta)}\right]. \tag{44}$$

This does not influence substantially the result of the estimate for the threshold number of PESW.

§5. CALCULATION OF THE CONTRIBUTION OF THREE-MAGNON PROCESSES

Three-magnon processes with transitions in the impurity are described by Hamiltonian terms of the form

$$H_3=\frac{i}{\mathcal{N}^{3/2}}\sum_{j\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}\left\{\{F^{(1)}_{j\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}b_{\mathbf{k}_1}b_{\mathbf{k}_2}b_{\mathbf{k}_3}{}^+a_j{}^+ + F^{(2)}_{j\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}b_{\mathbf{k}_1}b^+_{\mathbf{k}_2}b_{\mathbf{k}_3}{}^+a_j{}^+ + \text{H.c.}\right\}. \tag{45}$$

The amplitudes $F^{(1,2)}_{j\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}$ in these terms are given by

$$F^{(1)}_{j\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}=\frac{1}{8}\left(\frac{1}{2S}\right)^{1/2}\left(\frac{\omega_E{}^3}{\omega_{\mathbf{k}_1}\omega_{\mathbf{k}_2}\omega_{\mathbf{k}_3}}\right)(\cos\varphi_1\Lambda^{(1)}_{xx}+\cos\varphi_2\Lambda^{(2)}_{xx}$$
$$-\cos\theta_1\cos\varphi_1\Lambda^{(1)}_{yy}+\Lambda^{(2)}_{yy}\cos\theta_2\cos\varphi_2-i\Lambda^{(1)}_{yy}\cos\varphi_1-i\Lambda^{(2)}_{yy}\cos\varphi_2$$
$$-i\cos\theta_1\sin\varphi_1\Lambda^{(1)}_{xx}-i\cos\theta_2\sin\varphi_2\Lambda^{(2)}_{xx})\exp\{i(\mathbf{k}_1+\mathbf{k}_2-\mathbf{k}_3)\mathbf{R}_j\}, \tag{46}$$

$$F^{(2)}_{j\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3}=\frac{1}{8}\left(\frac{1}{2S}\right)^{1/2}\left(\frac{\omega_E{}^3}{\omega_{\mathbf{k}_1}\omega_{\mathbf{k}_2}\omega_{\mathbf{k}_3}}\right)(\cos\varphi_1\Lambda^{(1)}_{xx}+\cos\varphi_2\Lambda^{(2)}_{xx}$$
$$+\cos\theta_1\cos\varphi_1\Lambda^{(1)}_{yy}-\cos\theta_2\cos\varphi_2\Lambda^{(2)}_{yy}-i\cos\varphi_1\Lambda^{(1)}_{yy}-i\cos\varphi_2\Lambda^{(2)}_{yy}$$
$$+i\cos\theta_1\sin\varphi_1\Lambda^{(1)}_{xx}+i\cos\theta_2\sin\varphi_2\Lambda^{(2)}_{xx})\exp\{i(\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3)\mathbf{R}_j\}. \tag{47}$$

Relaxation of PESW is the result of two processes: coalescence of two PESW into a single secondary spin wave with transition of the impurity from the ground to the excited state, or with transition of the impurity from the excited to the ground state. Because of these processes (and their inverses), a change takes place in the spin-wave density in the $\mathbf{k}$-space regions near the resonance sphere $\omega_{\mathbf{k}} = \omega_p \mp \Omega_0$. A consistent analysis, similar to that in §4, shows that in the region $\omega_{\mathbf{k}} = \omega_p + \Omega_0$ the spin-wave density decreases in comparison with the thermal equilibrium level, whereas the number of secondary spin waves having $\omega_{\mathbf{k}} = \omega_p - \Omega_0$ increases sharply. Therefore, when studying nonlinear damping of PESW we can confine ourselves to the first of the foregoing processes.

Solving in the steady state the system of kinetic equations for the population differences of the impurities and for the spin-wave density, we obtain in full analogy with §4 the following results.

The average population difference of the ferromagnetic impurity ion levels changes with changing number ξ_0 of the PESW like

$$\sigma=\sigma_0, \quad \xi_0<\xi_c'; \qquad \sigma=\sigma_0(\xi_c'/\xi_0)^2, \quad \xi_0\geqslant\xi_c'. \tag{48}$$

The number of secondary spin waves concentrated near the sphere $\omega_{\mathbf{k}} = \omega_p - \Omega_0$ in $\mathbf{k}$-space is

$$\xi_1'=0, \quad \xi_0<\xi_c'; \tag{49}$$

$$\xi_1'=\frac{c\gamma_\parallel}{2\Gamma_s}\sigma_0\left[1-\left(\frac{\xi_c'}{\xi_0}\right)^2\right], \quad \xi_0>\xi_c'. \tag{50}$$

Thus, the three-magnon process leads to heating of the impurity subsystem when the number of the PESW exceeds a threshold value

$$\xi_c'=(\Gamma_s/2R'c\sigma_0)^{1/2}, \tag{51}$$

where

$$R'=(2\pi/\hbar)^2(|F^{(1)}|^2/\varkappa v_A), \tag{52}$$

and v_A is the velocity of spin wave of frequency $\omega_{\mathbf{k}} = \omega_p - \Omega_0$. Because of this process, an additional contribution appears in the PESW damping:

$$\Delta\Gamma_k'=0, \quad \xi_0<\xi_c'; \qquad \Delta\Gamma_k'=\frac{c\gamma_\parallel}{2\xi_0}\sigma_0\left[1-\left(\frac{\xi_c'}{\xi_0}\right)^2\right], \quad \xi_0>\xi_c'. \tag{53}$$

The maximum of $\Delta\Gamma_k'$ is reached at $\xi_0 = 3^{1/2}\xi_c'$ and amounts to

$$(\Delta\Gamma_k')^{max}=c\gamma_\parallel\sigma_0/\cdot 3^{3/2}\xi_c. \tag{54}$$

An estimate of the threshold number of the PESW for the three-magnon process shows that it exceeds the threshold value for the two-magnon process and is, under the same conditions, of the order of $\xi_c \sim 10^{-6}$–10^{-7}. It must be noted, however, as can be seen from (46), that to realize a two-magnon inelastic process it is necessary that the equilibrium directions of the effective impurity spin and of the spins of the host-lattice atoms be unequal (or that an anisotropic exchange interaction be present). In the case of a three-magnon process there are no such restrictions. We emphasize also that for the two-magnon process to take place it is necessary that the distance between the lower levels of the impurity ions satisfy the condition $\Omega_0 \leqslant \omega_p/2 - \omega_0$, where ω_0 is the minimum possible spin-wave frequency corresponding to $k = 0$. For a three-magnon process the corresponding condition takes the form $\Omega_0 \leqslant \omega_p - \omega_0$.

Thus, allowance for the different spin-wave relaxation on paramagnetic impurities is most important both for the determination of the threshold of the parametric excitation of the spin waves in experiments on parallel pumping in antiferromagnets, and also in the investigation of the above-threshold nonlinear behavior of the spin system in this situation.

In conclusion it must be noted that the principal effects discussed in the present article, particularly the phenomenon of "slow" relaxation, should be observed also in the study of the relaxation of phonons in antiferromagnetic crystals containing paramagnetic impurities.

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[1)]We describe here spin waves in classical fashion. In the calculation of linear relaxation on account of a one-magnon process the quantum and classical descriptions yield identical results, and the effects of nonlinear relaxation become substantial at high spin-wave intensities, when the classical description is known to applicable.

[2)]To satisfy this condition it is necessary to have $\xi_0 \ll \Theta/|\Phi|$.

[3)]Expression (25) was obtained by the method described in a preceding article.[8] The corresponding expression given earlier in Ref. 8 did not include the last two terms in the curly brackets. Equation (25) is more accurate; under typical experimental conditions, the correction, however, does not exceed 10%.

[4)]This choice is perfectly natural—we are interested in that solution which corresponds in the absence of parametric spin waves to an equilibrium thermal distribution.

[5)]Just as above, we leave out of (40) small terms proportional to p_1.

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