

# Quantum theory of spin-wave relaxation by two-level impurities

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We construct a complete microscopic theory of spin-wave relaxation due to modulation, by these waves, of the distances between the levels of the impurity paramagnetic ions in ferro- and antiferromagnets. The nonlinear effects produced when intense spin waves are excited in a crystal are calculated, and the possibility of cooling the impurity subsystem when intense spin waves interact with a field is indicated.

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Ferro- and antiferromagnetic crystals frequently contain as impurities ions of rare earth elements or of transition metals of the iron group. These ions have spin and orbital momenta and are characterized by a strong spin-orbit coupling. The influence of the crystal lattice field and the exchange interaction with the spins of the neighboring atoms, in conjunction with the spin-orbit interaction, leads to splitting of the energy levels of similar impurity ions. As a result, besides elastic scattering of the electron spin waves (SW), processes accompanied by transitions between levels in paramagnetic impurity ions become possible. The largest contribution to the SW relaxation is made by resonant transitions between levels, accompanied by absorption of one SW. These processes, however, are allowed only if the distance between the two lower levels in the impurity ion equals the frequency of the given spin wave. If this condition is not satisfied, the main contribution to the SW relaxation is due to the "slow" (longitudinal) relaxation mechanism,<sup>1</sup> which is caused by the modulation of the distance between the lower levels in the impurity ions by the spin wave. The purpose of the present paper is to construct a complete microscopic (linear and nonlinear) theory of slow relaxation in ferro- and antiferromagnets.<sup>1</sup>

## §1. FORMULATION OF MODEL

Under typical experimental conditions, the impurity density is low (tenths or hundredths of one percent). It is therefore natural to regard the impurity subsystem as an ensemble of independent ions located at random sites  $\{R_j\}$  of the crystal lattice. We assume also that the impurity atoms are two-level. Since the level population decreases sharply at low temperature with increasing level energy, one should expect the contributions from the third- and higher-order levels to be negligible under these conditions.<sup>2</sup>

We introduce the operators  $a_j^*$  and  $a_j$ , which relate the ground  $|0_j\rangle$  and excited  $|1_j\rangle$  states of an impurity ion located at the site  $R_j$  of the crystal lattice:

$$a_j^+|0_j\rangle=|1_j\rangle, \quad a_j|1_j\rangle=|0_j\rangle, \quad a_j^+|1_j\rangle=a_j|0_j\rangle=0. \quad (1)$$

The operators  $a_j^*$  and  $a_j$  pertaining to the same site satisfy Fermi permutation relations; the operators pertaining to different sites commute with each other.

Changing over to the SW variables for the spins of the host lattice and using the operators introduced above for the impurity ions, we can obtain (see the Appendix) the

following expression for the contribution made to the Hamiltonian and describing the modulation of the distance between the levels in the impurity ion

$$H=H_0+\delta H, \quad H_0=\sum_j \hbar\Omega_0 a_j^+ a_j, \quad (2)$$

$$\delta H=\mathcal{N}^{-1/2} \sum_{jk} (\Phi_{jk} b_k + \Phi_{jk}^* b_k^*) a_j^+ a_j,$$

where  $b_k^*$  and  $b_k$  are the canonical complex amplitudes of the spin waves,<sup>3</sup>  $\mathcal{N}$  is the total number of sites in the crystal, and  $\Phi_{jk}$  are the interaction amplitudes and are given in the Appendix.

We note that since  $\delta H$  commutes with  $H_0$ , the modulation of the distance between level is by itself incapable of producing transitions between these levels. It is therefore important to take into account the interaction of the impurity ions with the thermostat (the thermal phonons and magnons), an interaction that ensures relaxation of the populations in the impurity ions to the equilibrium value. We write down the Hamiltonian of this interaction in the general form

$$V=\sum_j V_j, \quad V_j=a_j F_j^+ + a_j^+ F_j, \quad (3)$$

so as to be able to replace in the final expressions the thermostat operators  $F_j^+$  and  $F_j$  by those concrete combinations of phonon or magnon creation and annihilation operators that correspond to different relaxation mechanisms of the impurity-ion population.

Owing to the interaction  $\delta H$ , the average spin-wave intensity  $n_k=\langle b_k^* b_k \rangle$  varies with time like

$$\dot{n}_k=-(2/\hbar\mathcal{N}^{1/2}) \sum_j \text{Im}(\Phi_{jk} \langle b_k a_j^+ a_j \rangle). \quad (4)$$

We shall see below that the correlator  $\langle b_k a_j^+ a_j \rangle$  is proportional to  $n_k$ , so that expression (4) determines the rate of the SW relaxation on account of the modulation of the distance between the levels and of the impurity ions.

In the calculation of the correlator  $\langle b_k a_j^+ a_j \rangle$  it can be taken into account that the characteristic SW relaxation times exceed by several orders the relaxation time of the level population in the impurity ions (it amounts to  $10^{-8}$ – $10^{-9}$  sec at low temperatures), and the adiabatic approximation can be used. That is to say, it can be assumed that the probability distribution for the impurity ions (i.e., their density matrix) attunes itself at each instant of time to the instantaneous value of the

spin-wave intensity [or to its slow amplitude, defined as  $\bar{b}_k = b_k \exp(i\omega_k t)$ ]. As a result, the correlator  $\langle b_k a_j^* a_j \rangle$  can be written in the form

$$\langle b_k a_j^* a_j \rangle = \int \text{Sp}(b_k a_j^* a_j, \sigma) P(\{b_k^*, b_k\}) d^2 b_k, \quad (5)$$

where the trace is taken over the atomic variables,  $P = P(\{b_k^*, b_k\})$  is the probability distribution for the complex SW amplitudes, while the density matrix  $\sigma$ , which depends on the spin-wave amplitudes  $\{\bar{b}_k\}$ , characterizes the atomic probability distribution that sets in at the specified values of these amplitudes.

## §2. THE CONTROL EQUATION

To find the density matrix  $\sigma$  of the impurity ions we use the approach developed by Lax (see Ref. 3). We note that owing to the low density of the impurities, they can be considered independently and it suffices therefore to consider one impurity atom with the distance between its levels modulated by the given field of spin waves with amplitudes  $b_k = \bar{b}_k \exp(i\omega_k t)$ , and take into account its interaction with the thermostat. The total Hamiltonian of such a system is

$$H_s = H + V + R, \quad (6)$$

where  $H$  is defined in (2),  $V$  is given by (3), and  $R$  is the Hamiltonian of the thermostat.

The equation of motion for the total density matrix  $\rho$  of this system is of the form<sup>4)</sup>

$$\partial \rho / \partial t = (H + V + R, \rho). \quad (7)$$

The density matrix  $\rho$  can be represented in the form

$$\rho = \sigma f_0 + \Delta \rho, \quad (8)$$

where  $\sigma = \text{Tr}_R \rho$  is the atomic density matrix (the trace is taken over the thermostat variables) and  $f_0$  is the equilibrium Gibbs density matrix of the thermostat at the temperature  $\theta$ ,

$$f_0 = \frac{\exp(-R/\theta)}{\text{Sp} \exp(-R/\theta)}. \quad (9)$$

We confine ourselves to those ion-relaxation mechanisms for which the operators  $F^*$  and  $F$  in (3) satisfy the condition

$$\text{Sp}_R (F f_0) = \text{Sp}_R (F^* f_0) = 0. \quad (10)$$

Taking in (7) the trace over the thermostat variables and using the condition (10), we obtain an equation for the atomic density matrix

$$\partial \sigma / \partial t = (H, \sigma) + \text{Sp}_R (V, \Delta \rho) + \text{Sp}_R (R, \rho). \quad (11)$$

Multiplying (11) by  $f_0$  and subtracting the obtained equation from (7), we can obtain an analogous equation for  $\Delta \rho$ , solve it in first order in the interaction  $V$ , and substitute the obtained value of  $\Delta \rho$  in (11). The final form of equation for the reduced density matrix  $\sigma$  (of the control equation) is then

$$\begin{aligned} \partial \sigma / \partial t = & (H, \sigma) + \int_{-\infty}^t dt' \text{Sp}_R (V, (V_0[t', t], \sigma[t', t'] f_0)) \\ & + \int_{-\infty}^t dt' \int_{t'}^t d\tau \text{Sp}_R (V, ((\delta H(\tau), V_0[t', t]), \sigma[t', t'] f_0)) \\ & - \frac{1}{2\hbar^2} \int_{-\infty}^t dt' \int_{t'}^t d\tau_1 \int_{t'}^{\tau_1} d\tau_2 \text{Sp}_R (V, (\delta H(\tau_1) \delta H(\tau_2) V_0[t', t], \sigma[t', t'] f_0)) \\ & - \frac{1}{2\hbar^2} \int_{-\infty}^t dt' \int_{t'}^t d\tau_1 \int_{t'}^{\tau_1} d\tau_2 \text{Sp}_R (V, (V_0[t', t] \delta H(\tau_1) \delta H(\tau_2), \sigma[t', t'] f_0)) + \dots \end{aligned} \quad (12)$$

We have used here the notation

$$V_0[t', t] = \exp \left\{ -\frac{i}{\hbar} (t-t') (H_0 + R) \right\} V \exp \left\{ \frac{i}{\hbar} (t-t') (H_0 + R) \right\}, \quad (13)$$

$$\sigma[t, t'] = \exp \left\{ -\frac{i}{\hbar} \int_{t'}^t H(\tau) d\tau \right\} \sigma(t') \exp \left\{ \frac{i}{\hbar} \int_{t'}^t H(\tau) d\tau \right\} \quad (14)$$

and carried out the expansion

$$\begin{aligned} & \exp \left[ \frac{i}{\hbar} \int_{t'}^t H(\tau) d\tau \right] \\ & = \exp \left[ \frac{i}{\hbar} H_0 (t-t') \right] \left\{ 1 - \frac{i}{\hbar} \int_{t'}^t \delta H(\tau) d\tau + \dots \right\}. \end{aligned} \quad (15)$$

Assuming the modulation of the distance between the atomic levels to be small,<sup>5)</sup> we have retained in (12) only terms up to second order in  $\delta H$ .

We are interested in that solution of (12) which is established in a given modulating spin-wave field. Were there no modulation the density matrix  $\sigma$  would relax to the Gibbs equilibrium form

$$\sigma_0 = \frac{\exp(-H_0/\theta)}{\text{Sp} \exp(-H_0/\theta)}, \quad (16)$$

or, taking into account the concrete form of the Hamiltonian  $H_0$  and the Fermi permutation relations for  $a^*$  and  $a$ ,

$$\sigma_0 = (e^{-\hbar \omega_0 / \theta} + 1)^{-1} [1 + (e^{-\hbar \omega_0 / \theta} - 1) a^* a]. \quad (17)$$

Since we assume the modulation amplitude to be small (see footnote 5), we can expect the density matrix  $\sigma$  in the steady state and in the presence of the modulating field of the SW to be given by the expansion

$$\sigma = \sigma_0 + \sum_k (\sigma_k^{(1)} e^{-i\omega_k t} + \text{H.c.}) + \dots, \quad (18)$$

which is a series in powers of the modulation amplitudes  $\{\bar{b}_k\}$ . The correction  $\sigma_k^{(1)}$  is proportional to the first power of the amplitudes  $\bar{b}_k$ , while the next terms, left out of (18), are of higher order in these amplitudes.

To find  $\sigma_k^{(1)}$  and the higher-order corrections we can use an iteration procedure. Indeed, substituting the expansion (18) in the control equation (12) and equating terms of equal order in the SW amplitudes, we obtain a system of equations for these quantities. To determine the first corrections  $\sigma_k^{(1)}$  it suffices to know the equilibrium density matrix  $\sigma_0$ , the second-order corrections call for additional knowledge of the first-order corrections, and so on. It is useful to note, when solving these equations, that the modulation of the distance between levels does not lead to the appearance of off-diagonal elements in the atomic density matrix,<sup>6)</sup> i. e., each of the corrections  $\Delta \sigma$  is of the form  $\Delta \sigma = \alpha + \beta a^* a$ , where  $\alpha$  and  $\beta$  are numerical coefficients. Since all the corrections of this form, as well as the equilibrium density matrix  $\sigma_0$ , commute with the Hamiltonian  $H(t)$ , the equality  $\sigma[t, t'] = \sigma(t')$  is satisfied for the steady state [see Eq. (14)].

## §3. LINEAR RESPONSE

The equation for finding the first-order correction  $\sigma_k^{(1)}$ , which is the linear response of the atomic system

to the modulating field of a spin wave with amplitude  $\bar{b}_k$ , is the following:

$$-i\omega_k \sigma_k^{(1)} = \int_{-\infty}^t dt' S_{PR}(V, (V_0[t', t], \sigma_k^{(1)}(t')) e^{i\omega_k(t-t')} + i(\Phi_k/\mathcal{N}^{1/2} \omega_k) \bar{b}_k \int_{-\infty}^t dt' [1 - e^{i\omega_k(t-t')}] S_{PR}(V, ((a^* a, V_0[t', t]), \sigma_{of0})). \quad (19)$$

The first term in the right-hand side of this equation describes the relaxation of the correction  $\Delta\sigma = \sigma_k^{(1)} \times \exp(-i\omega_k t)$  in the absence of a modulating field. It is therefore of the order of  $\gamma \sigma_k^{(1)}$ , where  $\gamma$  is the atomic frequency of the relaxation, amounting to  $10^8 - 10^9 \text{ sec}^{-1}$  at low temperatures, and is therefore much lower than the typical frequencies ( $\omega_k \sim 10^{11} \text{ sec}^{-1}$ ) excited in experiments on parallel pumping of spin waves. The indicated term is therefore small compared with the term  $-i\omega_k \sigma_k^{(1)}$  in the same equation, and can be neglected.

With the aid of the correction  $\sigma_k^{(1)}$  determined by (19) we can calculate the frequency of the linear SW relaxation due to modulation of the distance between the levels in the impurity bands. Knowing  $\sigma_k^{(1)}$ , we can calculate the contribution proportional to the intensity of the spin wave  $n_k$  made to the correlator  $\langle b_k a_j^* a_j \rangle$  and find, according to (4), an expression for the spin-wave relaxation frequency  $\Gamma_k^0$ :

$$\Gamma_k^0 = (c|\Phi|^2/\hbar^2 \omega_k^2) \{ \gamma_{II}(\Omega_0 + \omega_k) (I(\Omega_0) - I(\Omega_0 + \omega_k)) + \gamma_{II}(|\Omega_0 - \omega_k|) (I(\Omega_0 - \omega_k) - I(\Omega_0)) \}. \quad (20)$$

Here  $c$  is the dimensionless density of the impurity (the ratio of the number of impurity atoms to the total number  $\mathcal{N}$  of sites in the crystal),  $I(\Omega) = [\exp(\hbar\Omega/\theta) + 1]^{-1}$  is the average equilibrium population of the upper level in a two-level atom with distance  $\hbar\Omega$  between the levels at a temperature  $\theta$ .

The relaxation  $\Gamma_k^0$  is expressed in terms of the atomic population relaxation frequency  $\gamma_{II}(\Omega)$  for an atom with a distance  $\hbar\Omega$  between levels, as given by

$$\gamma_{II}(\Omega) = \hbar^{-2} \int_{\dot{}} du \{ \langle \langle F^+(0) F(u) \rangle \rangle + \langle \langle F(u) F^+(0) \rangle \rangle \} e^{i\Omega u + c.c.}, \quad (21)$$

where

$$F(u) = e^{(i/\hbar)Hu} F e^{-(i/\hbar)Hu}, \\ \langle \langle F^+(0) F(u) \rangle \rangle = S_{PR}(F^+(0) F(u) f_0).$$

The atomic relaxation frequencies were calculated earlier in Refs. 4 and 5 (see also Ref. 1). According to the results there, the main contribution to  $\gamma_{II}(\Omega)$  at temperatures down to several dozen degrees are made by transitions between impurity-atom levels with emission of one SW or of a phonon ("direct" relaxation processes). The expressions for  $\gamma_{II}(\Omega)$  in the case of various direct processes in ferro- and antiferromagnets are the following.

a) Transition with phonon emission

$$\gamma_{II}^{\text{ph}}(\Omega) = 3\pi^2 (\hbar/\rho v a^2) (|\Psi^{\text{ph}}|/\Theta_D)^2 \Omega (\Omega/\omega_D)^2 \text{cth}(\hbar\Omega/2\theta). \quad (21a)$$

Here  $\rho$  is the crystal density,  $v$  the phonon velocity,  $a$  the interatomic distance, and  $\Theta_D = \hbar\omega_D$  the Debye temperature.

b) Transitions with emission of spin wave in a ferro-

magnet:

$$\gamma_{II}^{\text{m}}(\Omega) = \pi^2 (\Psi^{\text{m}}/\Theta_C)^2 \hbar^{-1} [\hbar(\Omega - \omega_0)\Theta_C]^{1/2} \text{cth}(\hbar\Omega/2\theta). \quad (21b)$$

Here  $\Theta_C = \hbar\alpha/a^2$  is the Curie temperature, and the spin-wave dispersion law is taken in the form  $\omega_k = \omega_0 + \alpha k^2$ .

c) Transitions with spin-wave emission in an antiferromagnet of the "easy plane" type:

$$\gamma_{II}^{\text{m}}(\Omega) = \pi^2 (\Psi^{\text{m}}/\hbar\omega_E)^2 (\Omega/\omega_E) (\Omega^2 - \omega_0^2)^{1/2} \text{cth}(\hbar\Omega/2\theta). \quad (21c)$$

The SW dispersion law is taken in the form  $\omega_k^2 = \omega_0^2 + \omega_E^2 (ak)^2$ .

The values of the coefficients  $\Psi$  are given in the Appendix.  $\gamma_{II}^{\text{m}}(\Omega) = 0$  in formulas (21b) and (21c) if  $\Omega < \omega_0$ .

It is seen from expressions (21) that the dependence of the atomic relaxation  $\gamma_{II}$  on the temperature is the same in all cases, but the frequency dependences of  $\gamma_{II}(\Omega)$  are different. In particular, the contribution made to the atomic relaxation frequency by transitions with spin-wave emission vanishes when the distance between the levels in the impurity ion is less than the lowest frequency  $\omega_0$  of the spin waves in the crystal.

Formula (20) contains atomic relaxation frequencies taken at the combination frequencies  $\Omega = \Omega_0 \pm \omega_k$ . If the modulating frequency  $\omega_k$  is low compared with  $\Omega_0$ , then by expanding the expression in the curly brackets in (20) in terms of  $\omega_k$  we can arrive at the known result obtained for slow relaxation in the semiphenomenological theory.<sup>2</sup> The general expression obtained by us for  $\Gamma_k^0$  describes slow SW relaxation at any ratio of  $\Omega_0$  and  $\omega_k$  in the entire range of temperatures.

At high temperatures, when  $\theta/\hbar \gg \Omega_0$ ,  $|\Omega_0 - \omega_k|$ ,  $\Omega_0 + \omega_k$ , the value of  $\Gamma_k^0$  decreases with rising temperature like  $\Gamma_k^0 = A + B\theta^{-2}$ . In the opposite limit of low temperatures, the dependence of the SW relaxation frequency  $\Gamma_k^0$  on the temperature is different for the cases  $\Omega_0 > \omega_k$  and  $\Omega_0 < \omega_k$ :

$$\Gamma_k^0 \propto \exp[-(\hbar/\theta)(\Omega_0 - \omega_k)], \quad \Omega_0 > \omega_k, \\ \Gamma_k^0 \propto 1 - \exp[-(\hbar/\theta)(\omega_k - \Omega_0)], \quad \Omega_0 < \omega_k. \quad (22)$$

Thus, at  $\Omega_0 < \omega_k$  the relaxation  $\Gamma_k^0$  remains finite in the limit as  $\theta \rightarrow 0$ .

An estimate of the absolute value of  $\Gamma_k^0$  for a ferromagnetic yttrium iron garnet at  $\theta = 10 \text{ K}$ ,  $\Omega_0 = 10^{12} \text{ sec}^{-1}$ ,

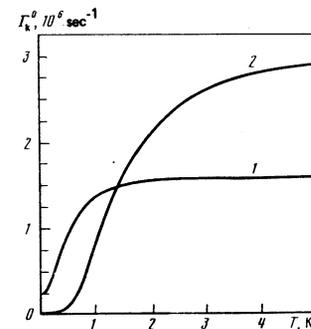


FIG. 1. Dependence of the relaxation  $\Gamma_k^0$  on the temperature for two values of the magnetic field  $H = 1 \text{ kOe}$  (1) and  $H = 4 \text{ kOe}$  (2), corresponding respectively to the conditions  $\Omega_0 < \omega_k$  and  $\Omega_0 > \omega_k$ .

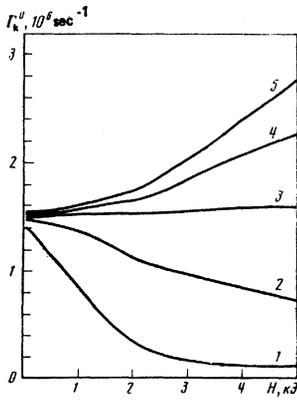


FIG. 2. Dependence of  $\Gamma_k^0$  on the magnetic field  $H$  for the temperatures (in K) 0.5 (1), 1 (2), 1.5 (3), 2 (4), and 2.5 (5).

$\omega_k = 10^{11} \text{ sec}^{-1}$ ,  $\gamma_{II} = 10^9 \text{ sec}^{-1}$ ,  $c = 1\%$ , and  $\Phi = 10^{-15} \text{ erg}$  yields  $\Gamma_k^0 = 10^8 \text{ sec}^{-1}$ , which agrees with the results of the experimental investigations (see Ref. 1).

Typical temperature dependences of  $\Gamma_k^0$  for antiferromagnets at helium temperatures are shown in Fig. 1. All the plots presented in this paper were calculated from the theoretical formulas at the parameter values  $c = 0.01\%$ ,  $|\Phi| = 10^{-14} \text{ erg}$ ,  $\Psi^{ph} = 10^{-7} \text{ erg/cm}$ ,  $\Psi^m = 10^{-15} \text{ erg}$ , and  $\omega_k = \omega_p/2 = 2.2 \times 10^{11} \text{ sec}^{-1}$ . The SW dispersion law was chosen in the form  $\omega_k^2 = \omega_0^2 + \omega_B^2 (ak)^2$ , where  $\omega_B = 10^{13} \text{ sec}^{-1}$ ,  $a = 10^{-8} \text{ cm}$ ,  $\omega_0^2 = g^2 [H(H + H_D) + \alpha/T]$ ,  $H_D = 4.4 \text{ kOe}$ , and  $\alpha = 6 \text{ K} \cdot \text{kOe}^2$ . The crystal density  $\rho$  is  $5 \text{ g/cm}^3$ , the phonon velocity  $v = 10^5 \text{ cm/sec}$ , and the Debye temperature  $\Theta_D = 10^{-14} \text{ erg}$ .

The distance between the levels in the impurity ions depends on the static magnetic field  $H$  and its orientation relative to the crystallographic axes. Figure 2 shows the relaxation frequencies  $\Gamma_k^0$  calculated from (20) and (21) as functions of the magnetic field for an antiferromagnet.<sup>7)</sup>

#### §4. NONLINEAR EFFECTS

The nonlinear effects are determined by corrections of higher order to the equilibrium atomic density matrix  $\sigma_0$ . The most important are: a) the time-independent second-order correction  $\Delta\sigma_0 \sim \sum_k b_k^* b_k$ , which yields the change of the average stationary population of the atomic levels in the presence of excited SW in the crystal; b) the third-order correction  $\Delta\sigma_k^{(1)} \sim (\sum_k b_k^* b_k) b_k \exp(-i\omega_k t)$  and the correction that is its Hermitian conjugate, which make it possible to calculate the nonlinear SW damping. To calculate these corrections we can use the iteration procedure described above. It is facilitated somewhat by noting that for each of the corrections it suffices to allow for the contributions that have the lowest order in the amplitude of the interaction of the impurity field with the thermostat.

A detailed calculation shows that the change of the stationary average population  $n = \langle a^\dagger a \rangle$  of the upper atomic level following excitation of  $N_0$  spin waves of frequency  $\omega_k = \omega_p/2$  in the crystal is given by

$$\Delta n = (4|\Phi|/\hbar\omega_p)^2 (N_0/\mathcal{N}) \gamma_{II}^{-1}(\Omega_0) \left\{ \gamma_{II}(|\Omega_0 - \omega_p/2|) \times [I(\Omega_0 - \omega_p/2) - I(\Omega_0)] - \gamma_{II}(\Omega_0 + \frac{\omega_p}{2}) [I(\Omega_0) - I(\Omega_0 + \omega_p/2)] \right\}, \quad (23)$$

where  $\mathcal{N}$  is the total number of sites in the crystal, and  $\Delta n = n - I(\Omega_0)$ .

Formula (23) can be represented in the form

$$\gamma_{II}(\Omega_0) [n - I(\Omega_0)] + (2|\Phi|/\hbar\omega_k)^2 (N_0/\mathcal{N}) \times \gamma_{II}(|\Omega_0 - \omega_k|) [I(\Omega_0) - I(\Omega_0 - \omega_k)] + (2|\Phi|/\hbar\omega_k)^2 (N_0/\mathcal{N}) \gamma_{II}(\Omega_0 + \omega_k) [I(\Omega_0) - I(\Omega_0 + \omega_k)] = 0. \quad (24)$$

Thus, from the quantum-mechanical point of view, an atom placed in a modulating SW field of frequency  $\omega_k = \omega_p/2$  spends, as it were, a fraction (proportional to the modulation depth) of its time in states with shifted distances between the ground and first excited levels, i. e., with shifted frequencies  $\Omega = \Omega_0 \pm \omega_k$ . The equilibrium values of the population of the upper level  $I(\Omega)$  are different for the unshifted ( $\Omega = \Omega_0$ ) and the two shifted positions of this level. The relaxation processes, tending to restore the thermal equilibrium disturbed by the level shifts, give rise to transitions within the impurity atoms, and the frequency of these transitions, i. e., the relaxation rate  $\gamma_{II}(\Omega)$ , depends on the distance  $\Omega$  between the levels.

When the temperature is relative high, so that  $\theta/\hbar \gg \Omega_0, |\Omega_0 - \omega_k|, \Omega_0 + \omega_k$ , the expansion  $I(\Omega_0 \pm \omega_k) \approx I(\Omega_0) \pm (\hbar\omega_k/2\theta)$  is valid, and expression (23) for the correction  $\Delta n = n - I(\Omega_0)$  takes the simpler form

$$\Delta n \approx (4|\Phi|/\hbar\omega_p)^2 (N_0/\mathcal{N}) (\hbar\omega_p/4\theta) \times \gamma_{II}^{-1}(\Omega_0) [\gamma_{II}(|\Omega_0 - \omega_p/2|) - \gamma_{II}(\Omega_0 + \omega_p/2)]. \quad (25)$$

As follows from the expressions (21a) and (21c) for  $\gamma_{II}(\Omega)$  in antiferromagnets, the relaxation rate is in this case always a monotonically increasing function of  $\Omega$ .<sup>8)</sup> Therefore the change of the average population is then always negative, i. e., the excitation of SW in the crystal is accompanied by an effective cooling of the impurity subsystem (the absolute value of the correction  $\Delta n$  decreases with increasing temperature like  $\theta^{-1}$ ). Since the atomic-relaxation frequency is higher when the level is shifted upward, the average population deviates towards the equilibrium population of the upward-shifted level, and is thus decreased.

In the opposite limit of low temperatures, the condition satisfied is  $I(\Omega_0 - \omega_k) \gg I(\Omega_0), I(\Omega_0 + \omega_k)$ , so that the following holds true:

$$\Delta n \approx (4|\Phi|/\hbar\omega_p)^2 (N_0/\mathcal{N}) \gamma_{II}^{-1}(\Omega_0) \gamma_{II}(|\Omega_0 - \omega_p/2|) I(\Omega_0 - \omega_p/2) \quad (26)$$

and heating of the impurity subsystem is therefore observed. Typical plots of  $\Delta n(\theta)$  for an antiferromagnet in a wide temperature interval are shown in Fig. 3.

We emphasize that in the situation considered by us the impurity subsystem is in a state of flux equilibrium—it is open and an energy flux passes through it from an intense external source (parametrically excited waves) into a thermostat. The possibilities of a transition of open systems with flux equilibrium into a more ordered state with increasing energy flux through the system (the "self-organization" phenomenon) are being extensively discussed in the literature (see, e. g., Haken's monograph.<sup>7)</sup> Included in this group of pheno-

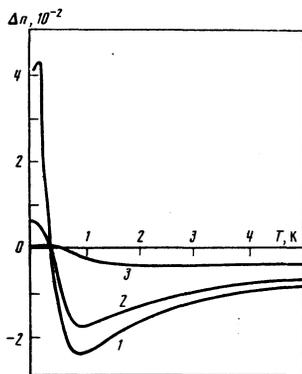


FIG. 3. Transition from heating to cooling of impurity subsystem with increasing temperature, at different values of the magnetic field (in kOe): 1) 1, 2) 1.5, 3) 4. The number of parametrically excited SW is fixed at  $N_0/N = 10^{-6}$ .

mena is also the effect observed by us, the cooling of impurities following parametric excitation of SW. In this effect an intense energy flux from parametric SW, passing through the impurity subsystem, draws additionally some power from the latter.

The change of the level populations in the impurity ions is reflected in a change of the SW relaxation frequency. If the crystal contains  $N_0$  parametrically excited spin waves with frequency  $\omega_p/2$  the SW relaxation  $\Gamma_k$  acquires on top of its equilibrium value  $\Gamma_k^0$  a correction  $\Delta\Gamma_k$  proportional to the number  $N_0$  of the excited spin waves<sup>9)</sup>:

$$\Delta\Gamma_k = c(|\Phi|^2/\hbar\omega_k)^2 [\gamma_{II}(\Omega_0 + \omega_k) - \gamma_{II}(|\Omega_0 - \omega_k|)] \Delta n, \quad (27)$$

where  $\Delta n$  is given by formula (23).

Since  $\Delta\Gamma_k \sim (N_0/N)$ , it is convenient to introduce a coefficient  $\eta$  of nonlinear damping of the excited spin waves, defined by the relation  $\Gamma_k = \Gamma_k^0 + \eta(N_0/N)$ . When (23) and (27) are taken into account, the expression for this coefficient takes the form

$$\eta = c(4|\Phi|^2/\hbar\omega_p)^2 \gamma_{II}^{-1}(\Omega_0) [\gamma_{II}(\Omega_0 + \omega_p/2) - \gamma_{II}(|\Omega_0 - \omega_p/2|)] \times \{ \gamma_{II}(|\Omega_0 - \omega_p/2|) [I(\Omega_0 - \omega_p/2) - I(\Omega_0)] - \gamma_{II}(\Omega_0 + \omega_p/2) [I(\Omega_0) - I(\Omega_0 + \omega_p/2)] \}. \quad (28)$$

In the high-temperature limit, when  $\theta/\hbar \gg \Omega_0$ ,  $|\Omega_0 - \omega_p/2|$ ,  $\Omega_0 + \omega_p/2$ , the coefficient  $\eta$  is negative and

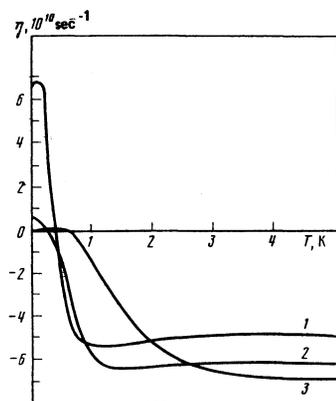


FIG. 4. Dependence of nonlinear-damping coefficient  $\eta$  on temperature at various  $H$  (in kOe): 1) 1.2, 2) 2.0, 3) 5.2.

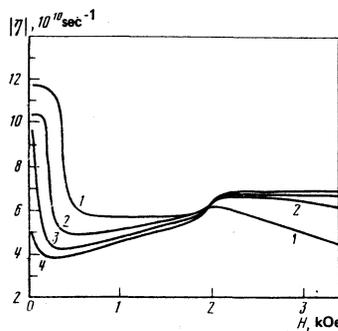


FIG. 5. Dependence of  $|\eta|$  on the magnetic field  $H$  at different temperatures (in K): 1) 1.0, 2) 1.5, 3) 2.0, 4) 2.5.

tends to a temperature-independent constant value ( $\eta$  is negative because in this case  $\Delta n < 0$ ). On the other hand, in the opposite low-temperature limit the nonlinear damping is positive, for heating of the impurity subsystem is then observed. Typical temperature dependences of the nonlinear damping  $\eta$ , calculated in accord with (28) for an antiferromagnet of the "easy plane" type, are shown in Fig. 4, while plots of  $|\eta|$  vs the magnetic field are shown in Fig. 5.

## 6. CONCLUSION

The SW relaxation mechanism considered by us and connected with the modulation of the level spacing in impurity paramagnetic ions by the spin waves exhibits significant peculiarities compared with other SW relaxation mechanisms. Since the interaction  $\delta H$  commutes with the Hamiltonian  $H_0$ , it does not by itself lead to transitions between stationary states of the impurity ion in any order of perturbation theory. The appearance of a contribution to the SW relaxation from the level modulation in the impurity ions is therefore due in principle to the finite widths of these levels or, in other words, to the relaxation of their populations. In the semiphenomenological theory,<sup>2</sup> the modulating action of the spin waves is taken into account within the framework of the kinetic equation for the average populations  $n$  of the impurity levels. This theory is therefore valid only in the case of smooth modulation, when the modulating frequency (i.e., the SW frequency  $\omega_k$ ) is low compared with the level spacing  $\Omega_0$  (only such slow external fields can be included in the kinetic equation). From the point of view of the semiphenomenological theory, the SW relaxation is then due to the delay between the attuning of the atomic populations to the instantaneous value of the level spacing.

In contrast to the approach of Hartman-Boutron,<sup>2</sup> the complete microscopic theory developed in the present paper does not assume that the modulating field is smooth—only a small modulation depth is assumed (see footnote 5). In the development of the theory we have used methods devised in connection with the study of laser systems<sup>3</sup> and stemming directly from the equation for the system density matrix. It follows from our formula (20) for the equilibrium SW relaxation, this quantity is expressed in terms of the impurity density  $c$ , the interaction amplitude  $\Phi$ , and the atomic relaxation frequencies  $\gamma_{II}$ , the latter being taken at the combination

frequencies  $\Omega_0 \pm \omega_k$ , as if the level spacing in the impurity ion were not  $\Omega_0$  but  $\Omega_0 \pm \omega_k$ , a situation directly connected with the modulating action of the spin wave. Since the frequency dependence of the atomic relaxation plays in this case a rather important role, we have calculated anew (with certain refinements) the expressions for this relaxation, with account taken of the "direct" processes that are most substantial at low temperatures. We have discussed, in addition, the nonlinear effects connected with the restructuring of the state of the impurity subsystem following parametric excitation of intense spin waves.

The situation most thoroughly investigated to date is the one in ferromagnetic yttrium iron garnet. Experiments on ferromagnetic resonance and parallel pumping, performed with a controlled content of one of the rare-earth elements or of the transition metals of the iron group, show (see Refs. 1, 8, and 9) that at temperatures below 100 K, starting with an impurity density of several tenths of one percent, the "slow" relaxation (due to modulation of the level spacing) makes the main contribution to the SW relaxation frequency. Since it follows from calculations<sup>1</sup> that the distances between the lower impurity-ion levels are in this case much larger than the ferromagnetic-resonance frequency or half the pump frequency, these experiments are well described by the semiphenomenological theory.

The  $\text{MnCO}_3$  and  $\text{CsMnF}_3$  crystals used in experiments on parallel pumping in antiferromagnets contain appreciable amounts of paramagnetic-ion impurities (according to Ref. 6, the investigated crystal contained 0.35% Fe, 0.04% Ni, and 0.01–0.45 Co). In these experiments, the plot of the SW relaxation frequency (determined from the threshold of the parametric resonance) vs the static magnetic field has a sharp peak interpreted as the consequence of resonant transitions between the impurity levels, accompanied by absorption of a spin wave when half the pump frequency equals the impurity-ion level spacing (which depends on the magnetic-field intensity).<sup>6</sup> At different values of the magnetic field, the impurity level spacing in these experiments can be either larger or smaller than the frequency of the excited SW. Therefore only the complete microscopic theory of slow SW relaxation, proposed in this paper, is valid in this case.

A comparison of the dependence of our calculated slow relaxation on the temperature (Fig. 1) and on the magnetic field (Fig. 2) with the experimental curves of Refs. 6 and 10 (see, in particular, Fig. 8 of Ref. 10) shows that our plot duplicates both the qualitative form of these curves and the absolute value of the SW relaxation in the investigated temperature range. It must also be noted that the clearly pronounced maxima observed when the crystal orientation is varied<sup>11</sup> are typical of slow relaxation (see Ref. 1) and are attributed to the decrease of the impurity-level spacing at certain directions of the static magnetic field.

The nonlinear negative damping of parametrically excited waves, due to partial cooling of the impurity subsystem, may be one of the causes of the "hard" SW excitation observed in the experiments. As seen from

Figs. 4 and 5, the fraction  $\Delta\Gamma = -\eta N_0/N$  of the relaxation that drops out in antiferromagnets at small excesses above the resonance threshold ( $N_0/N = 10^{-6}$ ) is approximately ten percent of the total SW relaxation. The value of  $\Delta\Gamma$  increases with decreasing magnetic field and, as follows from Fig. 5, increases with decreasing temperature in the interval from 1 to 2.5 degrees in weak magnetic fields, in accord with the experimental data.<sup>10</sup> The experimentally observed abrupt increase of  $\Delta\Gamma$  when half the pump frequency becomes equal to the impurity-level spacing can be attributed to heating of the impurity subsystem by direct transitions with absorption of a parametrically excited spin wave. It is important to emphasize that according to our present results the coefficient of nonlinear damping should reverse sign at temperatures below 0.5 K. Experiments at such low temperatures might show the degree to which hard excitation of SW in antiferromagnets is connected with the slow-relaxation mechanism.

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## APPENDIX

The energy spectrum of paramagnetic impurity ions in ferro- and antiferromagnets is the result of the action of the crystal electric field and of the exchange interaction with the spins of the neighboring atoms of the host lattice. As a result, because the degeneracy of the lower multiplet is lifted, the distance between the ground and first excited levels of the impurity ion turns out to be quite small and close in magnitude to the SW frequencies in the crystal. For 3d ions, the unfilled electron shell that determines such energy levels is an external shell and the level splitting due to the effective electric field exceeds considerably the splitting due to the spin-orbit coupling and to the exchange interaction. For 4f ions, the unfilled electron shell is screened by 5s and 5p electrons, so that the splitting due to the effective crystal field is much weaker than the spin-orbit splitting and turns out to be close to the exchange splitting. The energy levels of paramagnetic impurity ions should be calculated separately in each case; this calculation is not the subject of the present paper (for  $\text{Yb}^{3+}$  impurity ions in dodecahedral sites of the yttrium-iron-garnet lattice this calculation was performed, for example, in Ref. 12).

The modulation of the distance between impurity levels by spin waves is of exchange origin. For spin waves much longer than the characteristic interatomic distances, one can start out in the case of a ferromagnet with the exchange Hamiltonian

$$H_{ex} = \sum_j S_j \hat{\Lambda} J_j \quad (29)$$

or for a two-sublattice antiferromagnet with the Hamiltonian

$$H_{ex} = \sum_j (S_j^{(1)} \hat{\Lambda}^{(1)} J_j + S_j^{(2)} \hat{\Lambda}^{(2)} J_j). \quad (30)$$

The summation in (29) and (30) is over the sites  $\{R_j\}$  in which are located impurity ions having magnetic moments  $J_j$ . In (29),  $S_j$  is the averaged magnetic moment

of the host-lattice atoms that are adjacent to the impurity, while in (30)  $S_j^{(1)}$  and  $S_j^{(2)}$  are the instantaneous average moments of neighboring ions belonging to the two antiferromagnet sublattices. The tensors  $\hat{\Lambda}$ ,  $\hat{\Lambda}^{(1)}$ , and  $\hat{\Lambda}^{(2)}$  describe the exchange (anisotropic) interaction.

Carrying out the Holstein-Primakoff transformation of  $S_j$  or of  $S_j^{(1)}$  and  $S_j^{(2)}$ , we can express the Hamiltonians (29) and (30) in terms of spin-wave variables  $b_k^*$  and  $b_k$  (confining ourselves in the case of an antiferromagnet to the waves of the lower branch of the spectrum), we obtain

$$H_{ex} = U_0(\{J_j\}) + \frac{1}{\sqrt{N}} \sum_{jka} (b_k \mathcal{F}_{jka} + \text{H.c.}) J_{ja} + \dots \quad (31)$$

The contribution  $V_0$ , which does not depend on the spin-wave variables, can be left out and regarded as accounted for in the calculation of the energy spectrum of the impurity ions. The coefficients  $\mathcal{F}_{jka}$  (where  $\alpha = x, y, z$ ) are certain combinations of the elements of the tensor  $\Lambda$  (or of  $\Lambda^{(1)}$  and  $\Lambda^{(2)}$ ), as well as of the coefficients of the Holstein-Primakoff transformation and of additional transformations made on changing over to the variables of the SW in the crystal.

The amplitudes  $\Phi_{jk}$  in the Hamiltonian  $\delta H$  that describes the modulation are determined by the expressions

$$\Phi_{jk} = \sum_{\alpha} \mathcal{F}_{jka} \langle \langle 1_j | J_{ja} | 1_j \rangle - | 0_j | J_{ja} | 0_j \rangle \rangle, \quad (32)$$

in which  $|0_j\rangle$  and  $|1_j\rangle$  are the ground and first excited states of the impurity ion located at the site  $R_j$ .

A successive calculation yields the following concrete form of the amplitudes  $\Phi_{jk}$  for a two-sublattice antiferromagnet:

$$\Phi_{jk} = -1/2 i (S/2)^{1/2} (\omega_B/\omega_k)^{1/2} (\sin \theta_1 \Lambda_{zz}^{(1)} - \sin \theta_2 \Lambda_{zz}^{(2)}) e^{ikR_j}, \quad (33)$$

where  $\theta_1$  and  $\theta_2$  are the angles between the direction of the magnetic moment of the impurity ions in the ground state (the  $z$  axis) and the magnetization vectors of the two sublattices. In the derivation of (33) it was assumed that the exchange tensors are diagonal ( $\Lambda_{\alpha\beta}^{(1,2)} = \Lambda_{\alpha\beta}^{(1,2)} \delta_{\alpha\beta}$ ).

In the case of a ferromagnet with anisotropic exchange interaction and the directions of the magnetic moment of the impurities does not agree with the direction of the magnetization of the principal sublattice ( $\theta$  is the angle between these two directions) we obtain

$$\Phi_{jk} = (S/2)^{1/2} (\Lambda_{zz} - i \cos \theta \Lambda_{yz} - i \sin \theta \Lambda_{zx}) e^{ikR_j}. \quad (34)$$

The  $z$  axis is directed here along the magnetization of the host lattice.

We note that the angles  $\theta$ ,  $\theta_1$ , and  $\theta_2$  depend in the general case on the direction and magnitude of the external static magnetic field. As a result, similar relations may hold also for the interaction amplitudes  $\Phi_{jk}$ .

The Hamiltonian corresponding to the direct processes—transitions between levels, with absorption or emission of a spin wave—is of the form

$$H_1 = \mathcal{A}^{-1/2} \sum_{jk} (\Psi_{jk}^m b_k a_j^+ + \text{H.c.}). \quad (35)$$

The amplitudes  $\Psi_{jk}^m$  of the direct processes are given by

$$\Psi_{jk}^m = \sum_{\alpha} \mathcal{F}'_{jka} \langle 1_j | J_{ja} | 0_j \rangle. \quad (36)$$

A successive calculation yields for a two-sublattice antiferromagnet

$$\begin{aligned} \Psi_{jk}^m = & 1/2 (S/8)^{1/2} (\omega_B/\omega_k)^{1/2} (\cos \varphi_1 \Lambda_{xx}^{(1)} + \cos \varphi_2 \Lambda_{xx}^{(2)} - \cos \theta_1 \cos \varphi_1 \Lambda_{yy}^{(1)} \\ & + \cos \theta_2 \cos \varphi_2 \Lambda_{yy}^{(2)} - i \cos \varphi_1 \Lambda_{yz}^{(1)} - i \cos \varphi_2 \Lambda_{yz}^{(2)} - i \cos \theta_1 \sin \varphi_1 \Lambda_{zx}^{(1)} \\ & - i \cos \theta_2 \sin \varphi_2 \Lambda_{zx}^{(2)}) e^{ikR_j}, \end{aligned} \quad (37)$$

where  $(\theta_1, \varphi_1)$  and  $(\theta_2, \varphi_2)$  are the Euler angles of the magnetizations of the two sublattices, in a coordinate frame whose  $z$  axis is directed along the magnetic moment of the impurities.

In the case of a ferromagnet, the expression for  $\Psi_{jk}^m$  is

$$\Psi_{jk}^m = (S/8)^{1/2} (\Lambda_{xx} - \cos \theta \Lambda_{yy} - \sin \theta \Lambda_{xz} - i \Lambda_{xy} - i \cos \theta \Lambda_{yz} - i \sin \theta \Lambda_{zx}) e^{ikR_j}. \quad (38)$$

The Hamiltonian that describes interlevel transitions with absorption or emission of an acoustic phonon can be represented in the form

$$H_1 = \mathcal{A}^{-1/2} \sum_{jq\lambda} (qa) (\hbar/2M\omega_{q\lambda})^{1/2} (\Psi_{jq\lambda}^{ph} d_{q\lambda} a_j^+ + \text{H.c.}), \quad (39)$$

where  $d_{q\lambda}$  is the annihilation operator for a phonon of frequency  $\omega_{q\lambda}$ , polarization  $\mathbf{e}_{q\lambda}$ , and wave vector  $\mathbf{q}$ . It is assumed for simplicity that all atoms have the same mass  $M$ .

There are two causes of such transitions. Excitation of a phonon initiates a periodic change of the distance from the impurity ion to its nearest neighbors from the host lattice, so that changes occur in the crystal field that acts on the ion and in the constant of the exchange interaction with the spins of the neighboring atoms. If the direct transitions with phonon emission are of exchange origin, then the amplitude  $\Psi_{jq\lambda}^{ph}$  is given by

$$\Psi_{jq\lambda}^{ph} = \sum_i i [(q\Delta_i)/(qa)] e_{q\lambda}^x e^{iqR_i/S_{j+i}} \frac{\partial \Lambda}{\partial x_i} \langle 1_j | J_j | 0_j \rangle. \quad (40)$$

The summation here is over the atoms that neighbor with the impurity, are located at the sites  $R_{j+i}$ , and have average spins  $S_{j+i}$ :  $\Delta_i = R_{j+i} - R_j$  is the equilibrium distance to the impurity ions. We have taken into account the fact that the exchange-interaction tensor depends on the distance between the spins [ $\hat{\Lambda} = \hat{\Lambda}(x)$ ]. In the estimates the derivative  $\partial \hat{\Lambda}/\partial x_i$  can be set equal to  $\Lambda a^{-1}$ , where  $a$  is the interatomic distance.

An expression for the amplitude  $\Psi^{ph}$  governed by the crystal field variations can be found in Ref. 5.

1) The existing semiphenomenological linear theory of slow relaxation in ferromagnets<sup>2</sup> is valid only in a situation where in the distance between the lower levels of the ion is much larger than the frequency of the spin waves.

2) In a number of cases the distance between two lower levels is much less than the distance to the levels that follow. This takes place, in particular, if the lower pair of levels is a Kramers doublet and the splitting in the crystal field greatly exceeds the exchange splitting (see Ref. 1).

- <sup>3)</sup> We use the classical description of the spin waves. The calculated linear damping of the SW are the same in the classical and quantum descriptions. The nonlinear effects (in particular, the nonlinear damping of the SW) are significant only above the threshold of the spin-wave resonance, when the excited waves are already intense enough and the classical description is certainly valid for them.
- <sup>4)</sup> We use the notation  $(\dots, \dots) \equiv (1/i\hbar) [\dots, \dots]$ .
- <sup>5)</sup> We assume satisfaction of the condition  $|\Phi|^2(N_0/\gamma) \ll \hbar\Omega_0$ , where  $N_0$  is the total number of parametrically excited spin waves.
- <sup>6)</sup> This can be easily verified by analyzing directly the structure of the control equation (12).
- <sup>7)</sup> Using the data given for  $\text{MnCO}_3$  in Ref. 6, we put in the calculations  $\Omega_0 = 0.62 (H[\text{kOe}] + 0.4) \times 10^{11} \text{ sec}^{-1}$ .
- <sup>8)</sup> In ferromagnets, the contribution  $\gamma_{\parallel}^{\text{ph}}(\Omega)$  contains a decreasing section.
- <sup>9)</sup> Excitation of intense SW of frequency  $\omega_p/2$  in a crystal changes the relaxation also of other SW with different frequencies.
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