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Nonlinear effects involving localized magnon modes in impure ferromagnetic insulators*

I. P. Fittipaldi
Departamento de Física, Universidade Federal de Pernambuco, 50.000 Recife, Brazil and Department of Physics, Temple University, Philadelphia, Pennsylvania 19122

S. M. Rezende
Departamento de Física, Universidade Federal de Pernambuco, 50.000 Recife, Brazil
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A simple theoretical treatment for the study of nonlinear processes involving impurities in ferromagnetic insulators is presented. In particular, the possibility of exciting ferromagnetic magnons with very large wave vectors is analyzed theoretically. The proposed excitation mechanism is analogous to the first-order Suhl instability and takes place via nonlinear processes involving localized modes. A rough estimate of the critical power of a far-infrared laser source necessary to attain the instability threshold is given. It is noted that the threshold value is within the power capabilities of present pulsed far-infrared lasers, suggesting that the process is experimentally feasible.

I. INTRODUCTION

During the last few years there has been increasing interest in studying the effect of impurities as useful probes of the properties of magnetic insulators. It is well known that, under certain conditions, the introduction of an impurity spin in a Heisenberg exchange Hamiltonian can give rise to impurity modes whose energies may lie above or within the spin-wave band. These energy states may be classified according to the symmetry elements of the crystal lattice and are the so-called localized and virtual modes, respectively. The conditions for the appearance of localized and virtual modes associated with impurities in magnetic insulators have been studied by many investigators.[1-10] The significant thermodynamic effects of the impurity modes occur at temperatures comparable to their excitation energies. So, in contrast to the localized[11] modes, the virtual modes contribute dominantly to the thermodynamics of the system at low temperatures, since they are usually excited at lower energies.

Thermodynamic properties of such impurity modes have been studied by various authors.[12,13] Moreover, the presence of the spin-wave impurity states in magnetic crystals containing impurities has also been observed by a large number of different experimental techniques, such as optical measurement, neutron diffraction, and nuclear magnetic resonance. A recent review with detailed description of these different experimental techniques has been provided by Cowley and Buyers.[14]

The nonlinear behavior of the spin-wave modes beyond a critical occupation number is a well-known phenomenon (which is manifested as an unstable growth in the spin-wave population). In particular, the transient growth of the spin-wave population resulting from three-magnon nonlinear processes is due to the dipolar interaction, and as was pointed out by Suhl,[15] gives rise to the subsidiary resonance effect, observed at high-power ferromagnetic resonance experiments. In this case, the uniform mode (with $\vec{k}=0$) is pumped by a strong transverse microwave field and beyond a critical value h_c, the spin-wave modes (with $\vec{k}\neq 0$) directly coupled to the pumped mode, grow parametrically causing instabilities. This excitation method of spin waves is frequently referred to as "perpendicular pumping" and allows one to excite magnon pairs with low wave vectors only. Another extremely versatile method, which was proposed by Morgenthaler[16] and Schlömann,[17] is the so-called "parallel-pumping" technique, where the rf field is applied parallel to the static field, instead of being perpendicular to it. The advantage of this method over the former one lies in the fact that spin waves can be parametrically excited with a wide range of $\vec{k}$ vector. However, it presents difficulties if $\vec{k}$ is very large.[18] The large-$\vec{k}$ magnons in three-dimensional ferromagnets cannot be excited by this technique, since the exchange frequency is often considerably higher than the microwave frequency. Over the past several years, both techniques have been extensively employed as a useful means for studying relaxation mechanisms of spin waves, and considerable progress in the understanding of the ordered magnetic state has thus been made.

The objective of the present paper is to present a simple theoretical study of nonlinear processes involving localized modes associated with impurities in impure ferromagnets insulators. Here we are concerned with first-order nonlinear processes originating in dipolar interaction, i.e., three interacting magnon modes such that one is a localized mode. In a preliminary report presented some time ago[19] certain simple aspects of the present problem were examined, and in particular, results for the case where the magnitudes of the host and impurity spins are the same, i.e. $S=S'$, were reported. In the present paper we discuss the problem in detail and include the case $S\neq S'$. In Sec. II, the Heisenberg ferromagnet with nearest-neighbor exchange, containing a single ferromagnetically coupled impurity, is treated in the spin-wave approximation. By the use of a canonical transformation, the Hamiltonian is recast in a diagonal form. Next, some general features of a single substitutional impurity problem are discussed, and an asymptotic expression for the spin-wave mode amplitude associated with s-like localized modes is derived. The vertex of the interaction between a localized mode and two spin waves is derived explicitly (Sec. III), restricting ourselves to s-like localized modes. The s-like modes are of particular interest since they are directly associated with the motion of the impurity spin. As is reasonable to expect, we find that total wave vectors need not be conserved in such processes.

The main purpose of the present paper is to investigate theoretically the possibility of localized magnons (excited by an electromagnetic pulse) producing magnons in a threshold process. For this purpose, in Sec. IV we deal with the two magnons and one localized s mode coupling via dipolar interaction and an electromagnetic pulse (in the perpendicular configuration) from a strong laser source which enables us to pump the localized s mode. Under certain conditions, the instability threshold value is obtained (Sec. IV), and a rough estimate of the laser source power for some types of impurities is presented in Sec. V. Finally, in the Appendix an order-of-magnitude estimate of the relaxation rate of the localized s mode, due to splitting into two magnons, is presented.

From energy-conservation considerations, we can see that such magnons as one produces in these processes must have high energies. Therefore, it is worth seeking a deeper understanding of this mechanism since it offers a possible method of exciting magnons with very large $\vec{k}$. It should be emphasized that in this paper we have restricted ourselves to looking at a particular impure simple-cubic Heisenberg ferromagnet. Thus, the present results cannot be directly applied to known ferromagnetic insulators. However, we are concerned here primarily with the type of information which can be anticipated, and should a need develop to examine a particular lattice structure, the work of the present paper could, in most cases, be readily generalized.

II. SOME GENERAL FEATURES OF A SINGLE-SUBSTITUTIONAL-IMPURITY PROBLEM IN FERROMAGNETS

In this section, some features of a single-substitutional-impurity problem in a Heisenberg ferromagnet will be discussed. The aim is to derive an asymptotic expression for the spin-wave mode amplitudes associated with s-like localized modes, which will be used in Sec. III to treat the long-range dipolar terms.

The impurity energy states in a Heisenberg ferromagnet containing a ferromagnetic impurity were first examined by Wolfram and Callaway,[1] Takeno,[2] and Li and Zhu.[3] The present account is therefore rather brief, and it contains only as much information as is required to make the present paper self-contained.

We shall start from the usual isotropic nearest-neighbor exchange Hamiltonian which contains a single-substituted ferromagnetically coupled impurity (characterized by spin S' and exchange integral $J'>0$), which can be conveniently written in the form

$$\mathcal{H}_e=\mathcal{H}_e^0+\mathcal{H}_e^1\,, \tag{2.1}$$

where $\mathcal{H}_e^0$ is the host Hamiltonian,

$$\mathcal{H}_e^0=-J\sum_j\sum_\Delta \vec{S}_j\cdot\vec{S}_{j+\Delta}\,, \tag{2.2}$$

and $\mathcal{H}_e^1$ is the impurity Hamiltonian,

$$\mathcal{H}_e^1=2(J\vec{S}_0-J'\vec{S}'_0)\cdot\sum_\Delta\vec{S}_\Delta\,. \tag{2.3}$$

Here, the subscript j for the lattice vector runs over all sites, and Δ is summed over all nearest neighbors; the impurity site labeled by $j=0$ is assumed to be the origin of the coordinate system.

The Bose operators a_j and $a_j^\dagger$ for the spin deviations can be introduced through the Holstein-Primakoff transformation,[20] according to the relations

$$S_j^+=(2S)^{1/2}(1-a_j^\dagger a_{j/2S})^{1/2}a_j\,, \tag{2.4a}$$

$$S_j^-=(2S)^{1/2}a_j^\dagger(1-a_j^\dagger a_{j/2S})^{1/2}\,, \tag{2.4b}$$

$$S_j^z=s-a_j^\dagger a_j\,, \tag{2.4c}$$

and

$$S_0'^+=(2S')^{1/2}(1-a_0^\dagger a_{0/2S'})^{1/2}a_0\,, \tag{2.5a}$$

$$S_0'^-=(2S')^{1/2}a_0^\dagger(1-a_0^\dagger a_{0/2S'})^{1/2}\,, \tag{2.5b}$$

$$S_0'^z=S'-a_0^\dagger a_0\,. \tag{2.5c}$$

Restricting ourselves to the linear spin-wave approximation, which is valid at low temperatures, the Hamiltonian (2.1) can be written in a quadratic form as follows:

$$\mathcal{H}_e^0 = J_S \sum_j \sum_\Delta (a_j^\dagger a_j + a_{j+\Delta}^\dagger a_{j+\Delta} - a_j^\dagger a_{j+\Delta} - a_{j+\Delta}^\dagger a_j), \tag{2.6a}$$

$$\mathcal{H}_e^1 = 2JSz(\alpha-1)a_0^\dagger a_0 + 2JS(\alpha\beta-1)\sum_\Delta a_\Delta^\dagger a_\Delta - 2JS(\alpha\beta^{1/2}-1)\sum_\Delta (a_0^\dagger a_\Delta + a_\Delta^\dagger a_0), \tag{2.6b}$$

where z is the number of nearest neighbors, and α and β are, respectively, the ratios of the impurity effective exchange and the impurity spin to those of the host atoms,

$$\alpha = J'/J, \quad \beta = S'/S. \tag{2.7}$$

The quadratic Hamiltonian (2.6) can be brought to diagonal form by a canonical transformation, so that one can separate out the elementary excitations of the spin system. This method has been used by other authors[6,8] to treat impurity problems. The difference here is that we treat the impurity and the lattice spins in the same manner. So, we transform the operators a_j to a new set of operators a_λ through the transformations

$$a_j^\dagger = \sum_\lambda \Gamma_\lambda(j) a_\lambda^\dagger; \quad a_j = \sum_\lambda \tilde{\Gamma}_\lambda(j) a_\lambda, \tag{2.8}$$

where $\tilde{\Gamma}$ denotes the complex conjugate of Γ. Here, the spin-wave mode amplitudes $\Gamma_\lambda(j)$'s satisfy the following orthonormality conditions:

$$\sum_j \Gamma_\lambda(j)\tilde{\Gamma}_{\lambda'}(j) = \delta_{\lambda\lambda'}, \tag{2.9a}$$

$$\sum_\lambda \Gamma_\lambda(j)\tilde{\Gamma}_\lambda(j') = \delta_{jj'}. \tag{2.9b}$$

The inverse transformations

$$a_\lambda^\dagger = \sum_j \tilde{\Gamma}_\lambda(j) a_j^\dagger, \qquad a_\lambda = \sum_j \Gamma_\lambda(j) a_j \tag{2.10}$$

define the creation ($a_\lambda^\dagger$) and annihilation (a_λ) operators associated with the λ mode with eigenvalue E_λ, which satisfy the Bose commutation relations

$$[a_\lambda, a_{\lambda'}^\dagger]_- = \delta_{\lambda\lambda'}, \qquad [a_\lambda, a_{\lambda'}]_- = [a_\lambda^\dagger, a_{\lambda'}^\dagger]_- = 0. \tag{2.11}$$

With the transformation (2.8), the Hamiltonian (2.1) takes the diagonal form

$$\mathcal{H}_e = \sum_\lambda E_\lambda a_\lambda^\dagger a_\lambda, \tag{2.12}$$

where E_λ characterize the excitation energies of the magnons and the coefficients, $\Gamma_\lambda(j)$, are determined by the secular equation

$$E_\lambda\Gamma_\lambda(j) - 2JSz\Gamma_\lambda(j) + 2JS\sum_\Delta \Gamma_\lambda(j+\Delta) = 2JSz(\alpha-1)\Gamma_\lambda(0)\delta_{j0} + 2JS(\alpha\beta-1)\times\sum_\Delta \Gamma_\lambda(\Delta)\delta_{j\Delta} - 2JS(\alpha\beta^{1/2}-1)\times\sum_\Delta[\Gamma_\lambda(\Delta)\delta_{j0} + \Gamma_\lambda(0)\delta_{j\Delta}], \tag{2.13}$$

which is identical to that obtained by Wolfram and Callaway.[1] (Note that the right-hand side of this equation reveals the destruction of the translational invariance of the system due to the presence of the impurity spin.)

In the particular case of a pure ferromagnet ($\alpha=1$ and $\beta=1$), the spin-wave mode amplitudes reduce to the usual plane-wave form

$$\Gamma_k(j) = N^{-1/2} e^{i\vec{k}\cdot\vec{r}_j}, \tag{2.14}$$

where N is the total number of spins, $\vec{r}_j$ is the lattice vector connecting the origin with the jth site, and $\vec{k}$ denotes the wave vector of the excitation. In this case the eigenvalue E_λ also reduces to the familiar spin-wave dispersion relation (in the absence of an external magnetic field),

$$E_k = 2JSz(1-\gamma_k), \tag{2.15}$$

where

$$\gamma_k = z^{-1}\sum_\Delta e^{i\vec{k}\cdot\vec{r}_\Delta}. \tag{2.16}$$

The impurity energy states of the problem are determined by the solution of the $z+1$ basic secular equations,

$$E_\lambda\Gamma_\lambda(0) - 2JSz\Gamma_\lambda(0) + 2JS\sum_\Delta\Gamma_\lambda(\Delta) = 2JSz(\alpha-1)\Gamma_\lambda(0) - 2JS(\alpha\beta^{1/2}-1)\sum_\Delta\Gamma_\lambda(\Delta), \tag{2.17a}$$

$$E_\lambda\Gamma_\lambda(\Delta) - 2JSz\Gamma_\lambda(\Delta) + 2JS\sum_{\Delta'}\Gamma_\lambda(\Delta+\Delta') = 2JS(\alpha\beta-1)\Gamma_\lambda(\Delta) - 2JS(\alpha\beta^{1/2}-1)\Gamma_\lambda(0), \tag{2.17b}$$

which are obtained from Eq. (2.13), putting $j=0$ and $j=\Delta$. The impurity energy states [solutions of the Eqs. (2.17)] due to a single ferromagnetically coupled impurity in a Heisenberg ferromagnet have been studies through different approaches by several authors.[1-5] It is found that these energy states are discrete levels that may lie within or outside the energy band of the host (virtual and localized states, respectively), which may be classified according to the symmetry elements of the crystal lattice. For instance, in a simple cubic lattice the site symmetry of the defect is O_h, and there exist three types of magnon impurity modes, s-like (representation[21] Γ_1), p-like (Γ_{15}), and d-like (Γ_{12}). Among these modes, we are particularly interested here in the s-like modes, which are directly associated with the motion of the impurity spin characterized by a large amplitude predominantly located at the impurity and thus excitable by external radiation. Although the energies of the s-like localized modes have been previously studied in some detail (e.g., Refs. 1, 2, and 4), we discuss this case explicitly for completeness of our analysis, as well as for presenting an explicit expression of the criteria for the appearance of this type of localized modes in a simple cubic lattice. We follow closely the approach introduced by Ishii et al.[6] to discuss the corresponding problem for an antiferromagnetically coupled impurity.

The solutions of the set of secular equations (2.17) may be derived if we assume

$$\Gamma_\lambda(j) = V_0(2JSz)^{-1}g(j,0;\epsilon_\lambda) + (2JSz)^{-1}\sum_{\Delta} V_{\Delta}g(j,\Delta;\epsilon_\lambda), \qquad (2.18)$$

where V_0 and V_Δ are coefficients associated with the impurity site and its neighbors, respectively, and $g(j,j',\epsilon_\lambda)$ is a dimensionless Green's function defined as follows:

$$g(j,j';\epsilon_\lambda) \equiv (2JSz)G(j,j';E_\lambda), \qquad (2.19)$$

where $G(j,j';E_\lambda)$ is the usual pure-host Green's function, i.e.,

$$G(j,j';E_\lambda) = N^{-1}\sum_{k}\frac{e^{i\vec{k}\cdot(\vec{r}_j-\vec{r}_{j'})}}{E_k - E_\lambda}. \qquad (2.20)$$

Here the k summation extends over the first Brillouin zone. The dimensionless energy ϵ_λ introduced in the Eq. (2.19) is defined by

$$\epsilon_\lambda = 2E_\lambda/E_m - 1, \qquad (2.21)$$

where E_m ($=4JSz$) is the maximum excitation energy of the spin-wave band. (Note that $\epsilon_\lambda = 1$ and $\epsilon_\lambda = -1$ correspond, respectively, to the energies at the top and the bottom of the spin-wave band.) Substituting Eq. (2.18) into Eqs. (2.17a) and (2.17b), we obtain the following set of $z+1$ secular equations (valid for all cases of cubic crystals) which determine the coefficients V_0 and V_Δ, and the eigenvalues ϵ_λ of the system:

$$V_0[1+(\alpha-1)g(0,0;\epsilon_\lambda) - (\alpha\beta^{1/2}-1)g(\Delta,0;\epsilon_\lambda)] + \Big(\sum_{\Delta'} V_{\Delta'}\Big)\Big((\alpha-1)g(\Delta,0;\epsilon_\lambda) - (\alpha\beta^{1/2}-1)z^{-1}\sum_{\Delta''} g(\Delta,\Delta'';\epsilon_\lambda)\Big) = 0\,; \qquad (2.22a)$$

$$V_0[-(\alpha\beta^{1/2}-1)z^{-1}g(0,0;\epsilon_\lambda) + (\alpha\beta-1)z^{-1}g(\Delta,0;\epsilon_\lambda)] + \sum_{\Delta'} V_{\Delta'}[\delta_{\Delta\Delta'} - (\alpha\beta^{1/2}-1)z^{-1}g(\Delta,0;\epsilon_\lambda) + (\alpha\beta-1)z^{-1}g(\Delta,\Delta';\epsilon_\lambda)] = 0\,. \qquad (2.22b)$$

In arriving at this equation, we have used the fact that, in cubic lattices, $g(\Delta,0;\epsilon_\lambda)$ and $\sum_{\Delta'} g(\Delta,\Delta';\epsilon_\lambda)$ are independent of the direction of the vector $\vec{r}_\Delta$. To solve the system of Eqs. (2.22a), and (2.22b), we shall confine ourselves to the solutions corresponding only to s-like modes which are characterized[6] by the site symmetry given by nonzero V_0 and $\sum_\Delta V_\Delta$. To pick up, from Eqs. (2.22), solutions with $V_0 \neq 0$ and $\sum_\Delta V_\Delta \neq 0$, we combine the Eq. (2.22a) with the following equation:

$$V_0[-(\alpha\beta^{1/2}-1)g(0,0;\epsilon_\lambda) + (\alpha\beta-1)g(\Delta,0;\epsilon_\lambda)] + \Big(\sum_{\Delta'} V_{\Delta'}\Big)\Big(1-(\alpha\beta^{1/2}-1)g(\Delta,0;\epsilon_\lambda) + (\alpha\beta-1)z^{-1}\sum_{\Delta''} g(\Delta,\Delta'';\epsilon_\lambda)\Big) = 0, \qquad (2.22c)$$

which is obtained summing Eq. (2.22b) over Δ. In this manner, the following relationship, which determines the energy of the s-like modes, is obtained:

$$D(\epsilon_s) = (\alpha+\alpha\beta-1) + (\alpha\beta-1)\epsilon_s + g(0,0;\epsilon_s)[(\alpha-1) + (\alpha+\alpha\beta-2)\epsilon_s + (\alpha\beta-1)\epsilon_s^2] = 0\,. \qquad (2.23)$$

In deriving the above we have used the following identity satisfied by the Green's functions[1,6]:

$$z^{-1}\sum_{\Delta} g(j,\Delta;\epsilon_\lambda) = -\delta_{j0} - \epsilon_\lambda g(j,0;\epsilon_\lambda)\,. \qquad (2.24)$$

Numerical analysis of the Eq. (2.23) has been presented by many authors.[1-4] The perfect-crystal Green's function that appear into Eq. (2.23) is known over the whole parameter ϵ_s range, and in our calculations we shall use the tabulated values for a simple cubic lattice given by Yussouff and Mahanty.[22] For our present purposes it is sufficient to restrict ourselves to solutions of Eq. (2.23) whose energies may appear above the top of the spin-wave band ($\epsilon_s > 1$); i.e., to the localized s-like modes. A criteria for the existence of these localized spin-wave modes can be derived. First, we note that $D(\epsilon_s)$ is a monotonically increasing function of ϵ_s, and that

$$\lim_{\epsilon_s\to\infty} D(\epsilon_s) = 1\,. \qquad (2.25)$$

Hence, we have at most one solution in the region $\epsilon_s > 1$ if

$$D(\epsilon_s = 1) < 0\,. \qquad (2.26)$$

From Eq. (2.23) we can see that this condition can be satisfied if

$$\alpha > \left(\frac{1}{2} + \frac{F(-1)-1}{2F(-1)-1}\beta\right)^{-1}, \qquad (2.27)$$

where $F(-1) = 1.51638\cdots$, defined by

$$F(n) = N^{-1}\sum_{k}(1-\gamma_k)^n\,, \qquad (2.28)$$

which has been evaluated by Watson.[23] The boundary curve in the α-β plane for the appearance of the s-like localized modes as well as isoenergy curves for these modes are shown in Fig. 1.

Now, we address ourselves to the task of obtaining the spin-wave mode amplitudes associated with s-like localized modes at the impurity and at its neighboring sites, as well as deriving an asymptotic expression for $\Gamma_s(j)$ with large r_j. Using the symmetry properties[2] of the s-mode ($V_0 \neq 0$; $V_1 = V_2 = \cdots V_z \equiv V_\Delta$) and Eq. (2.24), we can rewrite Eq. (2.18) as follows:

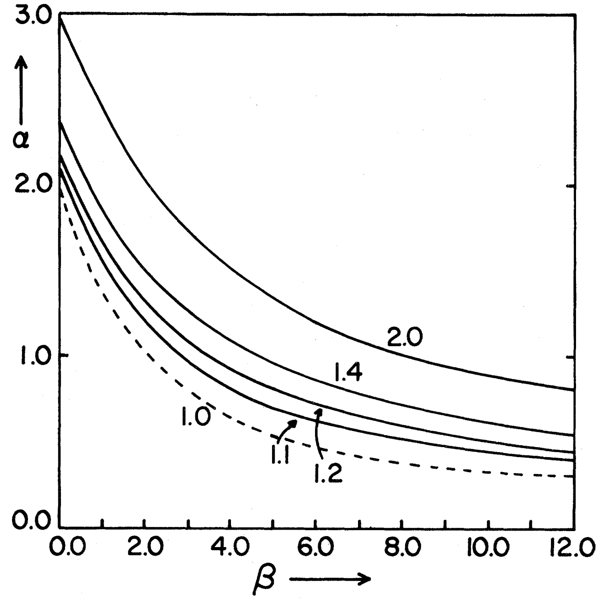

FIG. 1. Isoenergy curves for the s-like localized modes in the α-β plane, where $\alpha = J'/J$ and $\beta = S'/S$. The number associated with each curve denotes the dimensionless energy ϵ_s of the localized s mode. The dashed line shows the boundary curve which is obtained as a limiting case of Eq. (2.27). Localized s modes exist only in the regions above this boundary curve.

$$\Gamma_s(j) = \mu_s[-v_s\delta_{j0} + (1 - v_s\epsilon_s)g(j,0;\epsilon_s)]\,, \qquad (2.29)$$

where

$$\mu_s = V_0/2JSz\;;\quad v_s = zV_\Delta/V_0\,. \qquad (2.30)$$

The factor v_s can be determined from Eq. (2.22c) and the relation (2.24)

$$v_s = \frac{(\alpha\beta-1) + [(\alpha\beta^{1/2}-1) + (\alpha\beta-1)\epsilon_s]g(0,0;\epsilon_s)}{\alpha\beta^{1/2} + (\alpha\beta-1)\epsilon_s + [(\alpha\beta^{1/2}-1)\epsilon_s + (\alpha\beta-1)\epsilon_s^2]g(0,0;\epsilon_s)}\,, \qquad (2.31)$$

while the quantity μ_s can be obtained using the normalization condition (2.9a)

$$\mu_s = [v_s^2 - 2v_s(1 - v_s\epsilon_s)g(0,0;\epsilon_s) + (1 - v_s\epsilon_s)^2\mu(\epsilon_s)]^{-1/2}\,, \qquad (2.32)$$

where

$$\mu(\epsilon_s) = N^{-1}\sum_{k}(\epsilon_s + \gamma_k)^{-2}\,. \qquad (2.33)$$

In the region of $\epsilon_s > 1$ an approximate expression for $\mu(\epsilon_s)$ can be evaluated by expanding the denominator of the Eq. (2.33) in powers of γ_k/ϵ_s, which for a simple cubic lattice reads

$$\mu(\epsilon_s) = \sum_{n=0} c_n \epsilon_s^{-2(n+1)} , \tag{2.34}$$

where the first coefficients c_n are given by

$$\begin{aligned} c_0 &= 1 \\ c_1 &= 3[F(2) - 1] \\ c_2 &= 5[F(4) - 6F(2) + 5] \\ c_3 &= 7[F(6) - 15F(4) + 75F(2) - 61] \\ &\vdots \end{aligned} \tag{2.35}$$

The functions $F(2)$, $F(4)$, and $F(6)$ defined in the Eq. (2.28) have been calculated by Tahir-Kheli.[24] So, using Eqs. (2.29) and (2.32) the coefficients $\Gamma_s(j)$ at the impurity and at its neighboring sites are found.

$$|\Gamma_s(0)|^2 = v_s(1-\xi)^2/\eta , \tag{2.36a}$$

$$|\Gamma_s(\Delta)|^2 = v_s^2 \xi^2/\eta . \tag{2.36b}$$

Here

$$\xi = (1/v_s)(1 - v_s\epsilon_s) g(0,0;\epsilon_s) , \tag{2.37a}$$

$$\eta = v_s^2 - 2v_s(1 - v_s\epsilon_s) g(0,0;\epsilon_s) + (1 - v_s\epsilon_s)^2 \mu(\epsilon_s) . \tag{2.37b}$$

The spin-wave mode amplitude associated with an s-like mode at any site r_j from the impurity can be written as

$$\Gamma_s(j) = \left(\frac{v_s\epsilon_s - 1}{v_s - (1 - v_s\epsilon_s) g(0,0;\epsilon_s)} \right) \times \Gamma_s(0) g(j,0;\epsilon_s) , \quad j \neq 0 . \tag{2.38}$$

To determine the asymptotic form of $\Gamma_s(j)$ for large r_j, we use the asymptotic form of the Green's function $G(j, 0, E_s)$ for large values of r_j, which has been discussed in detail by Callaway[25] through application of the method of stationary phase. According to this author, and considering only s-like localized modes ($\epsilon_s > 1$), the dimensionless Green's function (2.19) has the following asymptotic form:

$$g(j,0;\epsilon_s) = \frac{3}{2}\frac{a}{\pi}\left(1 - \frac{(K_s a)^2}{10}\right)\frac{e^{-K_s r_j}}{r_j}, \quad r_j \gg 0 , \tag{2.39}$$

where a denotes the lattice constant of a sc lattice, and K_s is determined by the equation

$$\gamma_{k=iK_s} = \epsilon_s , \ \epsilon_s > 1 . \tag{2.40}$$

Thus, use of Eq. (2.39) allows us to express $\Gamma_s(j)$, for sites away from the impurity, in the form

$$\Gamma_s(j) = (a/4\pi)\Lambda(\epsilon_s)(e^{-K_s r_j}/r_j) , \tag{2.41}$$

where

$$\Lambda(\epsilon_s) = z\left(1 - \frac{(K_s a)^2}{10}\right) \times \left(\frac{v_s\epsilon_s - 1}{v_s - (1 - v_s\epsilon_s) g(0,0;\epsilon_s)}\right)\Gamma_s(0) . \tag{2.42}$$

[Note that, as expected, $\Gamma_s(j)$ has the expected damped form.] With the aim of studying explicitly the dependence of some quantities with the energy ϵ_s, we will write the inverse range parameter K_s as an approximate solution of Eq. (2.40), i.e.,

$$K_s = a^{-1}[z(\epsilon_s - 1)]^{1/2} , \tag{2.43}$$

which is valid for values of ϵ_s in the range $1 < \epsilon_s < 1.5$.

Before concluding this section, let us consider the correction to the energy of the s-like localized modes due to the presence of a static magnetic field. The shift of energy levels of a ferromagnet containing an impurity atom due to the magnetic field has been analyzed by Ishii *et al.*[6] and by Izyumov and Medvedev.[26] Here, we shall concentrate our attention on a first-order correction of the energy of the localized s-modes. In the spin-wave approximation, the exchange Hamiltonian of Eq. (2.1) must be supplemented by the field-dependent part,

$$\mathcal{H}_Z = \mathcal{H}_Z^0 + \mathcal{H}_Z^1 , \tag{2.44}$$

where $\mathcal{H}_Z^0$ is the Zeeman host Hamiltonian

$$\mathcal{H}_Z^0 = g\mu_B H \sum_j a_j^\dagger a_j , \tag{2.45a}$$

and $\mathcal{H}_Z^1$ accounts for the impurity

$$\mathcal{H}_Z^1 = g\mu_B(\delta - 1) H a_0^\dagger a_0 , \tag{2.45b}$$

where

$$\delta = g'/g . \tag{2.46}$$

Here, H is the magnetic field assumed to be along the z direction, μ_B is the Bohr magneton, and g and g' are the Landé factors of the host and the impurity atoms, respectively. Now, the expression (2.20) for the Green's function of an ideal crystal is still valid but with the energy of the spin wave E_k, replaced by

$$E_k(H) = 2JSz(1 - \gamma_k) + g\mu_B H . \tag{2.47}$$

To obtain the energy correction of s-like localized modes, we express the Hamiltonian given in (2.45a), and (2.45b) in terms of the operators a_λ through the canonical transformations (2.8) and pick-up the diagonal part in a_λ. In this manner, we obtain

$$E_s(H) = 2JSz[1+\epsilon_s(H)] + g\mu_B H\,, \tag{2.48}$$

$$\epsilon_s(H) = \epsilon_s^0 + \frac{g\mu_B H}{2JSz}(\delta - 1)|\Gamma_s(0)|^2\,, \tag{2.49}$$

where ϵ_s^0 is the dimensionless energy for the case of zero field. It is worth mentioning that (in contrast to p and d states) the energy shift of the s-like modes, besides the usual amount $g\mu_B H$, depends also upon the g factor of the impurity. Thus, to take into account the presence of the external magnetic field, the dimensionless energy ϵ_s that appears in the previous equations must be replaced by that given in Eq. (2.49). [Henceforth, for simplicity, the explicit indication of the dependence on the field H of $\epsilon_s(H)$ will be omitted.]

III. VERTEX OF THE INTERACTION

Akhiezer[27] was the first to show that the dipolar interaction in ferromagnetic crystals can induce transitions between spin waves. From this theoretical analysis, it has been possible to explain[15,28] the nonlinear behavior of the ferromagnetic resonance[29,30] at high rf magnetic fields. In this section we present the derivation of the vertex of the interaction between a localized s mode and two spin waves, which will be used in the remainder of the paper. The coupling between the localized mode and the magnons takes place via the dipolar interaction.

The whole Hamiltonian of the system under study, is now assumed to include, in addition to the exchange and Zeeman parts, the dipolar interaction

$$\mathcal{H}_d = \mathcal{H}_d^0 + \mathcal{H}_d^1\,, \tag{3.1}$$

where $\mathcal{H}_d^0$ is the dipolar host Hamiltonian,

$$\mathcal{H}_d^0 = \frac{1}{2}\sum_{i\neq j}(g\mu_B)^2 r_{ij}^{-3}[\vec{S}_i\cdot\vec{S}_j - 3(\vec{r}_{ij}\cdot\vec{S}_i)(\vec{r}_{ij}\cdot\vec{S}_j)r_{ij}^{-2}]\,, \tag{3.2a}$$

and $\mathcal{H}_d^1$ takes account of the presence of the impurity, i.e.,

$$\mathcal{H}_d^1 = (g\mu_B)^2\sum_{j(\neq 0)} r_{0j}^{-3}\{(\delta\vec{S}_0' - \vec{S}_0)\cdot\vec{S}_j - 3[\vec{r}_{0j}\cdot(\delta\vec{S}_0' - \vec{S}_0)][\vec{r}_{0j}\cdot\vec{S}_j]r_{0j}^{-2}\}\,. \tag{3.2b}$$

Here $\vec{r}_{ij}$ is the relative position vector connecting the ith and jth sites, and δ is defined in Eq. (2.46). The Hamiltonian (3.1) can be written as a sum of terms involving an increasing number of coupled spin-deviation operators using a higher-order expansion of the Holstein-Primakoff transformation given in Eqs. (2.4) and (2.5). We shall limit ourselves to the lowest-order terms of interest, i.e., those with three operators, since they give rise to the first-order nonlinear process, and therefore, are responsible for providing the lowest critical threshold. Thus, neglecting terms higher than the third order, $\mathcal{H}_d$ becomes

$$\begin{aligned}\mathcal{H}_d = &-(2S)^{1/2}\sum_{i\neq j}(F_{ij}a_j a_i^\dagger a_i + \tilde{F}_{ij}a_i^\dagger a_i a_j^\dagger)\\ &-(2S)^{1/2}(\delta-1)\sum_{j(\neq 0)}(F_{0j}a_j a_0^\dagger a_0 + \tilde{F}_{0j}a_0^\dagger a_0 a_j^\dagger)\\ &-(2S)^{1/2}\tfrac{1}{4}(\delta\alpha - 1)\sum_{j(\neq 0)}(F_{0j}a_j^\dagger a_j a_j + \tilde{F}_{0j}a_j^\dagger a_j^\dagger a_j)\\ &-(2S)^{1/2}(\delta\alpha^{1/2}-1)\sum_{j(\neq 0)}(F_{0j}a_0 a_j^\dagger a_j + \tilde{F}_{0j}a_j^\dagger a_j a_0^\dagger)\,,\end{aligned} \tag{3.3}$$

where

$$F_{ij} = -\tfrac{3}{2}(g\mu_B)^2 r_{ij}^{-3}[r_{ij}^z(r_{ij}^x - ir_{ij}^y)r_{ij}^{-2}]\,. \tag{3.4}$$

This Hamiltonian can now be expressed in terms of the operators a_λ, through the canonical transformations (2.8) which can be conveniently rewritten in such a way as to separate out the elementary excitations of the system

$$a_j = \sum_k \Gamma_k(j)a_k + \sum_\nu \Gamma_\nu(j)a_\nu\,, \tag{3.5}$$

where a_k is the destruction operator for the perturbed magnons of the host energy band, and a_ν is the localized magnon operator. Retaining only the terms which can destroy the localized modes, we obtain

$$\mathcal{H}_d^{\nu\to k+k'} = \sum_{k,k',\nu}[V(k,k';\nu)a_\nu a_k^\dagger a_{k'}^\dagger + \text{H.c.}]\,, \tag{3.6a}$$

where

$$V(k,k';\nu) = N^{-1}F(\nu)F(k')\sigma(k,k';K_\nu) \tag{3.6b}$$

and

$$\sigma(k,k';K_\nu) = \frac{a^{-2}}{|\vec{k}+\vec{k}'|^2 + K_\nu^2}\,, \tag{3.7}$$

$$K_{\nu=s} = a^{-1}(\sigma_0)^{-1/2}\,;\quad \sigma_0 = [z(\epsilon_s - 1)]^{-1}\,, \tag{3.8}$$

$$F(\nu = s) = -\Lambda(\epsilon_s)\,;\quad F(k) = (2S)^{1/2}\sum_j F_{ij}e^{i\vec{k}\cdot\vec{r}_{ij}}\,. \tag{3.9}$$

In arriving at these results, we have neglected the contributions of the last three terms of Eq.

(3.3) compared to that of the first term. The reason is that in the first term of Eq. (3.3), the sum runs over all sites i and j, whereas in the remaining terms the summation runs only over the sites j. On the other hand, we note that this result should be exact for the case of an impurity spin with $S'=S$, since only the first term of Eq. (3.3) survives. Furthermore, in writing the above results, we have used the asymptotic form for $\Gamma_s(j)$ given by Eq. (2.41), which is a good approximation for treating the long-range dipolar terms. (Note that for the perturbed spin waves, we assume plane wave form.)

Equations (3.6a) and (3.6b) represent a process in which a localized magnon decays into a pair of magnons of the host energy band (splitting process), as well as, the reverse process where two magnons of the host energy band (with different energies and momenta) are destroyed and a localized magnon is created (confluence process). Both of these processes are illustrated in Fig. 2. The presence of the function $\sigma(k,k';K_\nu)$ in the vertex[31] (3.6b) reveals that, as expected, wave vector need not be conserved in these processes. However, we note that the maximum amplitude of $\sigma(k,k';K_\nu)$ occurs for $\vec{k}=-\vec{k}'$, indicating that this is the dominant process. In Fig. 3, the function $\sigma(k,k';K_\nu)$ is plotted as a function of aq, where $q=|\vec{k}+\vec{k}'|$, for several representative values of the dimensionless energy ϵ_s. Examination of this figure reveals that when the energy of the localized mode appears near the top of the magnon energy band, the peak of the function $\sigma(k,k';K_\nu)$ at $\vec{k}=-\vec{k}'$ becomes more pronounced. In this situation, practically, there exist only processes in which $\vec{k}=-\vec{k}'$. Note that from a theoretical point of view it is possible to obtain localized modes with energies close to that of the spin-wave energy at the top of the band if a convenient selection of the impurity parameters α and β is available (see Fig. 1).

In order to study the nonlinear process of interest, it is desirable to rewrite the total Hamiltonian of the magnetic system in terms of new boson operators $c_\lambda^\dagger$ and c_λ, as follows:

$$\mathcal{H}_m=\sum_k \hbar\omega_k c_k^\dagger c_k+\sum_{\nu(=s)}\hbar\omega_\nu c_\nu^\dagger c_\nu$$

$$+\sum_{k,k',\nu}[\bar{V}(k,k';\nu)c_\nu c_k^\dagger c_{k'}^\dagger+\text{H.c.}], \qquad (3.10a)$$

where now

$$\bar{V}(k,k';\nu)=N^{-1}F(\nu)\bar{F}(k')\sigma(k,k';K_\nu), \qquad (3.10b)$$

with

$$\bar{F}(k)=F(k)u_k^2. \qquad (3.11)$$

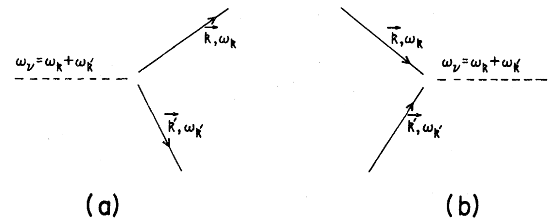

FIG. 2. Schematic representation of the first nonlinear processes involving a localized impurity mode (dashed line): (a) splitting process; (b) confluence process. Note that wave vector need not be conserved in these processes.

These new operators, c_λ's, are introduced by a Bogolyubov canonical transformation,[32]

$$a_k=u_k c_k-v_k^* c_{-k}^\dagger;$$
$$a_k^\dagger=u_k^* c_k^\dagger-v_k c_{-k}, \qquad (3.12)$$

which diagonalizes the quadratic part of the Hamiltonian of the pure ferromagnetic system including the contributions from Zeeman, exchange, and dipolar interactions. Here the transformation (3.12) has been applied to the magnon operators, but considered as an identity for the localized modes operators $a_s^\dagger$ and a_s. In fact, this means that we are neglecting the effects of the quadratic

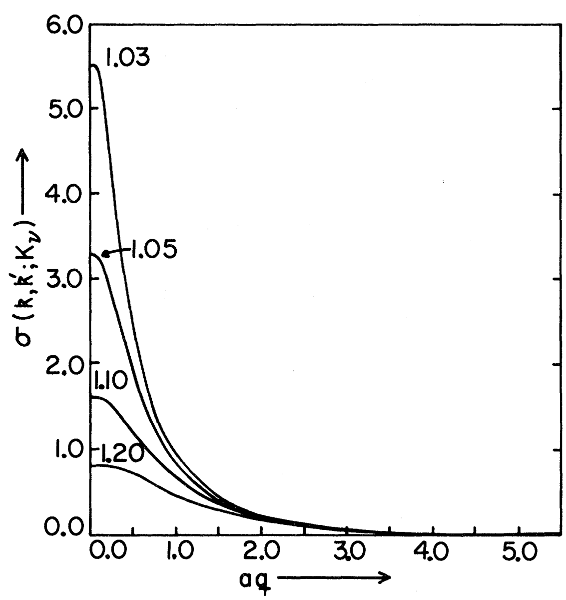

FIG. 3. Behavior of the function $\sigma(k,k';K_s)$ (see the definition in the text) as a function of aq, where $q=|\vec{k}+\vec{k}'|$. Numerical figures denote the values of the dimensionless energy ϵ_s of the s-like localized magnon modes.

part of the dipolar interaction on the energies of the localized modes, since it is very small compared to the corresponding part of the exchange interaction. The parameters u_k and v_k in Eq. (3.11) and Eq. (3.12) are defined in Ref. 33. In Eq. (3.10a), $\hbar\omega_s$ is the energy of the localized s mode and $\hbar\omega_k$ is the dispersion relation for magnons. Note that throughout this paper we have assumed that we are concerned with impurity crystals exhibiting a sufficiently small concentration of impurities, so that the effects of the magnetic impurities on the magnons of the host energy band can be neglected.

IV. PARAMETRIC EXCITATION OF SPIN WAVES VIA LOCALIZED MAGNON MODES

The experimental nonlinear excitation of magnons (i.e., perpendicular pumping) was discovered in connection with ferromagnetic resonance experiments.[29,30] In these experiments, a subsidiary resonance and a saturation of the main resonance were observed at high power levels. These effects were explained by Suhl[15,28] as arising from a transient growth of the spin-wave population involving three-magnon and four-magnon transitions, respectively. In particular, the first-order Suhl instability (three-magnon nonlinear process) is due to the dipolar interaction and occurs at lowest critical threshold. This mechanism has proved to be a very useful parametric excitation process, which has been employed for studying relaxation mechanisms of spin waves. In these experiments, from direct measurements of the critical field, it is possible to obtain information about coupling coefficients among spin waves, as well as their relaxation rates.

In the discussion of Sec. III, we concluded that the dipolar interaction in ferromagnetic crystals containing impurities can induce transitions between localized modes and host spin-waves modes. So, in analogy to the well-known first-order Suhl instability, if a localized mode is pumped by a strong enough electromagnetic field, one finds that beyond a critical value of the field, the spin-waves modes, directly coupled to the pumped localized mode, grow parametrically, causing instabilities. When this condition is satisfied, the growth rate from pumping exceeds the decay rate from the various relaxation processes. In what follows we shall deduce the critical number of photons, n_q^{crit}, at the threshold of the nonlinear process.

In order to study the resonance process, we take the Hamiltonian (3.10a), and write it in the form

$$\mathcal{H}_m = \sum_k \hbar\omega_k c_k^\dagger c_k + \sum_{\nu(=s)} \hbar\omega_\nu c_\nu^\dagger c_\nu + \sum_k \hbar[\phi(k;s)\, c_s\, c_k^\dagger c_{-k}^\dagger + \text{H.c.}], \qquad (4.1)$$

where we have considered only the processes in which $\vec{k} = -\vec{k}'$, since it produces the lowest threshold. In Eq. (4.1), $\phi(k;s)$ is the coupling coefficient which depends upon the impurity parameters

$$\phi(k;s) = N^{-1/2} f_s f_k, \qquad (4.2)$$

where

$$f_s = \sigma_0 F(\nu = s) \qquad (4.3)$$

and

$$f_k = N^{-1/2}\hbar^{-1} F(k) u_k^2. \qquad (4.4)$$

The threshold condition, which results from the third-order term in the Hamiltonian (4.1), can be obtained either from the equation of motion[34] or the instability criterion.[35] Using the fact that instability occurs when the number of quanta in the k mode required to maintain equilibrium becomes infinite, White and Sparks[35] have established a general instability criterion which can be applied to any boson-boson process. Using the results of these authors in our case (for a fuller account, the reader is referred to Ref. 35), we find that the critical number of the pumped localized mode n_s^{crit} at the threshold is given by

$$n_s^{\text{crit}} = \eta_k^2 / |\phi(k;s)|^2 . \qquad (4.5)$$

Here η_k denotes the relaxation rate of a potentially unstable k mode. [Note that the conservation of energy is required; i.e., $\hbar\omega_s = 2\hbar\omega_k$, with, of course, the restriction $\hbar\omega_s \leq 2\hbar\omega_m$ ($\hbar\omega_m \equiv E_m$).]

The external excitation is given by an electromagnetic pumping field, which is perpendicular to the static one. The perturbing Hamiltonian is given by

$$\mathcal{H}_{m-p}(t) = -g\mu_B \sum_{j(\neq 0)} h_x(t) S_j^x - g'\mu_B h_x(t) S_0'^x, \qquad (4.6)$$

where the magnetic field operator can be represented in the form

$$h_x(t) = i\left(\frac{2\pi\hbar\omega}{v_1}\right)^{1/2} (\vec{q}_0 \times \vec{e}_q) \times \left(b e^{-i[(\omega/c)y - \omega t]} - b^\dagger e^{i[(\omega/c)y - \omega t]}\right) \qquad (4.7a)$$

or

$$h_x(t) = h^+ e^{i\omega t} + h^- e^{-i\omega t}. \qquad (4.7b)$$

Here $b^\dagger$ and b are interpreted as creation and annihilation operators of photons. In Eq. (4.7a),

v_1 is the volume which defines, for n_q incident photons, an electromagnetic energy density $n_q\hbar\omega/v_1$, c is the velocity of light, and $\vec{e}_q$ is the polarization vector of the magnetic field. The wave vector of the electromagnetic field $\vec{q}$ ($|q|=\omega/c$; $\vec{q}_0=\vec{q}/|q|$) is parallel to the y axis. The interaction Hamiltonian (4.6) can be written in terms of the operators a_λ and $a_\lambda^\dagger$ using Eqs. (2.4), (2.5), and the canonical transformations (2.8). The relevant part of this Hamiltonian responsible for the pumping of the localized s modes, with $\hbar\omega=\hbar\omega_s$ is

$$\mathcal{H}_{s-p}=-i\hbar\gamma(bc_s^\dagger-\mathrm{H.c}), \tag{4.8}$$

where

$$\gamma=N^{-1/2}G\Gamma_s(0), \tag{4.9}$$

and

$$G=g'\mu_B\hbar^{-1}(NS'\pi\hbar\omega/v_1)^{1/2}. \tag{4.10}$$

In the above, $\Gamma_s(0)$ is the amplitude of the localized s mode at the impurity site [given in Eq. (2.36a)], and N is the number of host sites in a volume v of the crystal around the impurity. From Eq. (4.8) we can note that in the stationary linear region (see, for instance, Ref. 34) the occupation number of the localized magnons is proportional to the number of incident photons

$$n_s=(\gamma/\eta_s)^2 n_q. \tag{4.11}$$

Here η_s denotes the relaxation rate (introduced phenomenologically) of a localized s mode from the various possible relaxation processes. (In the Appendix, an order of magnitude estimate of η_s due to the process of splitting into two magnons will be presented.)

Using Eqs. (4.5) and (4.11) we get the critical number of photons at the threshold of the nonlinear processes with $\hbar\omega=\hbar\omega_s=2\hbar\omega_k$,

$$n_q^{\text{crit}}=(\eta_s\eta_k/\gamma|\phi(k;s)|)^2, \tag{4.12}$$

or, more explicitly,

$$n_q^{\text{crit}}=\frac{1}{m|\Gamma_s(0)|^2|f_s|^2}\left(\frac{\eta_s\eta_k}{G|f_k|}\right)^2, \tag{4.13}$$

where m is the concentration of impurities. As mentioned earlier, we are considering the case of small-impurity concentration only. Therefore, the effect of adding more impurity spins is assumed to be simply additive.[36]

The amplitude of the electromagnetic field applied parallel to the x axis [$h_x(t)=h\cos\omega t$] is found by equating the electromagnetic energy density to $n_q^{\text{crit}}\hbar\omega/v_1$. The result is

$$h^{\text{crit}}=\frac{1}{m^{1/2}|\Gamma_s(0)||f_s|}\left(\frac{\eta_s\eta_k}{R|f_k|}\right), \tag{4.14}$$

where

$$R=\tfrac{1}{4}\hbar^{-1}g'\mu_B(2NS')^{1/2}. \tag{4.15}$$

Note that after experimental determination of the physical parameter h^{crit}, it is possible to obtain information about coupling coefficients between localized s modes and spin waves, as well as, measurements of the relaxation rates of impurities. In Sec. V, a rough estimate of the critical power of a far-infrared laser source necessary to attain the instability threshold will be presented.

V. DISCUSSION

As explained in the preceding sections, when the resonance of a localized s mode is pumped above a certain threshold, magnon pairs of the host spin-wave energy band are driven unstable. As the localized magnon energies lie above the top of the spin-wave band ($\gtrsim 50$ cm^{-1}), energy conservation shows that the magnon pairs[37] produced by the decay are close to the edge of the Brillouin zone. This immediately suggests that the present process can be used to measure the lifetime of magnons in this region ($k\sim10^7-10^8$ cm^{-1}), analogous to the well-known experiments in the microwave range. The mechanism proposed here is represented schematically in Fig. 4.

To make an order-of-magnitude estimate of the power of a far-infrared laser source necessary to attain the critical photon number given in Eq. (4.13), let us take the following typical values for the parameters η_k and $|f_k|$; $\eta_k\simeq10^8$ sec^{-1}, $|f_k|\simeq1$ sec^{-1}, which are valid for yttrium iron garnet (YIG) and several simple antiferromagnets. For the impurity parameters we take $m=0.1\%$, $|\Gamma_s(0)|^2\simeq10^{-1}$, $|f_s|^2\simeq10^2$, and $\eta_s\simeq10^9$ sec^{-1}. The latter three numbers are valid, for instance, for im-

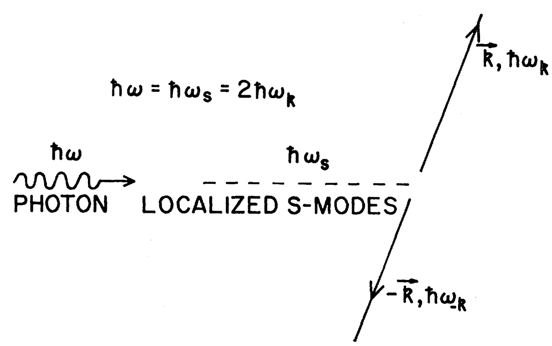

FIG. 4. Schematic representation of the mechanism proposed here to excite magnons with very large $\vec{k}$.

purities characterized by $S'=S$, $J'=1.53J$, and $S'=2S$, $J'=1.20J$. The order-of-magnitude computation of the relaxation rate of localized modes, presented in the Appendix, gives $\eta_s \simeq 10^8-10^9$ sec^{-1}. (It is obtained assuming the relaxation of the s modes to be due to the splitting process described earlier.) Using these numbers, we obtain

$$W_{\text{crit}}/\sigma \sim 10^3 - 10^4 \text{ W/cm}^2 , \qquad (5.1)$$

where σ is the sectional area of the beam. Therefore, for millimeter beams the critical power is of the order of tens of hundreds of watts, which is within the power capabilities of present pulsed far-infrared lasers. The difficulty in the experiments suggested here seems to be the same as those referring to the direct excitation of localized modes, namely, the tuning of the resonance to the existing laser transitions.

Finally, it is worth mentioning that although the calculations presented here apply directly to impurities in simple cubic ferromagnets, it is reasonable to expect that the qualitative features of the problem do not depend critically upon the crystal structure. We also expect that the order of magnitude of the results should be the same for antiferromagnets, in which localized modes have been investigated experimentally.

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APPENDIX: ESTIMATE OF RELAXATION DUE TO THE SPLITTING PROCESS

In this Appendix we present an order-of-magnitude calculation of the relaxation rate of a localized s mode due to splitting into two magnons of the host energy band. To proceed with this task, let us take the interaction term of Eq. (3.10a),

$$\mathcal{H}_d^{\nu \to k+k'} = \sum_{k,k',\nu} [\overline{V}(k,k';\nu)\, c_\nu c_k^\dagger c_{k'}^\dagger + \text{H.c.}] . \qquad \text{(A1)}$$

[The function $\overline{F}(k)$ is as defined in Eq. (3.11).] Let us rewrite this in the form

$$\overline{F}(k) = g\mu_B M_0 \phi(k) , \qquad \text{(A2)}$$

where $M_0=(N/V)g\mu_B S$ is the saturation magnetization and $\phi(k)\simeq 1$ (see Ref. 38). The probability that a localized magnon decays into a pair of magnons of the host energy band can be calculated with first-order perturbation theory. The nonvanishing matrix element of (A1) corresponding to this process [see Fig. 2(a)] is

$$\langle(\eta_\nu - 1), (n_k+1), (n_{k'}+1)|\mathcal{H}_d| n_\nu, n_k, n_{k'}\rangle = [n_\nu(n_k+1)(n_{k'}+1)]^{1/2}[\overline{V}(k,k';\nu)+\overline{V}(k',k;\nu)] . \qquad \text{(A3)}$$

Using perturbation-theory procedures, one can now find the transition probability for the decrease of one localized magnon mode due to the splitting process. If this is subtracted from the probability for the inverse process, one gets the rate equation, defining the rate of increase of the occupation number, i.e.,

$$\frac{dn_\nu}{dt} = \sum_{k,k'} [P_{\text{trans}}(n_\nu \to n_\nu - 1) - P_{\text{trans}}(n_\nu \to n_\nu + 1)] . \qquad \text{(A4)}$$

Using the golden rule, this gives

$$\frac{dn_\nu}{dt} = L_d(\{n\}) , \qquad \text{(A5)}$$

where

$$L_d(\{n\}) = \frac{1}{2}\frac{2\pi}{\hbar}\sum_{k,k'} |\overline{V}(k,k';\nu)+\overline{V}(k',k;\nu)|^2 \times [n_\nu(n_k+1)(n_{k'}+1) - (n_\nu+1)n_k n_{k'}] \times \delta(\hbar\omega_k + \hbar\omega_{k'} - \hbar\omega_\nu) . \qquad \text{(A6)}$$

Note that $L_d(\{\overline{n}\})=0$, where $\overline{n}=[\exp(\hbar\omega/k_B T)-1]^{-1}$ is the thermal equilibrium occupation number. So, the relaxation time τ_ν for this process is given by

$$\frac{1}{\tau_\nu} = \frac{\delta L_d(\{n\})}{\delta n_\nu} = \frac{\pi}{\hbar}\sum_{k,k'} |\overline{V}(k,k';\nu)+\overline{V}(k',k;\nu)|^2 \times (\overline{n}_k + \overline{n}_{k'} + 1)\,\delta(\hbar\omega_k + \hbar\omega_{k'} - \hbar\omega_\nu) , \qquad \text{(A7)}$$

where

$$(\bar{n}_k + \bar{n}_{k'} + 1) = \frac{1}{2}\,\frac{\sinh(\frac{1}{2}\hbar\omega_\nu/k_B T)}{\sinh(\frac{1}{2}\hbar\omega_k/k_B T)\sinh(\frac{1}{2}\hbar\omega_{k'}/k_B T)} \tag{A8a}$$

$$\simeq k_B T\,\frac{\omega_\nu}{\hbar\omega_k\omega_{k'}}, \quad k_B T \gg \hbar\omega_{k,k'}. \tag{A8b}$$

Using Eqs. (3.10b) and (A2), we obtain

$$\frac{1}{\tau_s} = \frac{4\pi}{\hbar}(g\mu_B M_0)^2\omega_s k_B T\,|F(\nu=s)|^2 N^{-2} \times \sum_{k,k'} \frac{\sigma^2(k,k';K_s)}{\hbar\omega_k\hbar\omega_{k'}}\,\delta(\omega_s-\omega_k-\omega_{k'}), \tag{A9}$$

which gives

$$\frac{1}{\tau_s} = \frac{4\pi}{\hbar^3}(g\mu_B M_0)^2\omega_s k_B T\,|F(\nu=s)|^2\, I(K_s), \tag{A10}$$

where

$$I(K_s) = \frac{v_c^2}{(2\pi)^6}\int\!\!\int \frac{d^3k\,d^3k'}{\omega_k\omega_{k'}} \times\sigma^2(k,k';K_s)\,\delta(\omega_s-\omega_k-\omega_{k'}). \tag{A11}$$

The integral above was calculated using a linear dispersion relation for magnons ($\hbar\omega_k = \hbar\omega_0 + Dk$). The result for the relaxation frequency ($\eta_s = 1/\tau_s$) is found to be

$$\eta_s = \frac{\pi}{16}\sigma_0 |F(\nu=s)|^2 \left(\frac{\hbar\omega_s}{\hbar\omega_m}\right)^2 \left(\frac{g\mu_B M_0}{\hbar\omega_m}\right)^2 \left(\frac{k_B T}{\hbar}\right), \tag{A12}$$

where σ_0 is defined in Eq. (3.8). To make an order-of-magnitude estimate, let us take the following typical values: $H \simeq 10^3$ Oe, $M_0 \simeq 10^3$ Oe, $T \simeq 100\,^\circ$K, $g \simeq 2$, $J \simeq 10^{-15}$–10^{-16} ergs. For the impurity parameters we assume $S' = S$, $J' = 1.53J$ and $S' = 2S$, $J' = 1.20J$. Using these numbers, we obtain

$$\eta_s \sim 10^8 - 10^9\ \text{sec}^{-1}. \tag{A13}$$

Finally, it is worth mentioning that the total relaxation frequency for a localized s mode is the sum of the contributions from all possible interaction processes. However, from a theoretical point of view, it is possible to obtain localized modes with energies close to the top of the spin-wave band in such way that the present process can be the most important relaxation mechanism.

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