γ, ε indicated in the figure the SW frequency is 0.010 below the gap edge.

The results above show that the penetration depth of low-frequency SW into the medium significantly exceeds the wave length for $k \ll 1$. This result can be obtained by methods of the theory of elasticity only for $k^2 \ll \gamma$. For $\gamma \lesssim k^2 \ll 1$, however, the dependence of this penetration length on the wave length becomes nonlinear. The high-frequency SW also has a large penetration depth for $k \ll 1$.

The polarization of low-frequency SW is primarily of bending nature of all k. The high-frequency polarization of SW is primarily along the x-axis.

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NOTATION

Here $\mathbf{u}(\mathbf{n})$ is the vector of atomic displacement from the equilibrium position at site $\mathbf{n}$; Φ_{ik} is the force constant matrix; a is the atomic separation in the base plane; ε_a is the interplanar atomic separation; α is the force constant for atoms of the same layer; $\gamma\alpha$ is the force constant for atoms of neighboring layers; k_1, k_2, k_3 are dimensionless wave vector components; ω is the dimensionless vibrational frequency; ω_s is the surface vibrational frequency.

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Relaxation of impurities in a nonresonant field and phonon amplification

M. I. Dykman

Institute of Semiconductors, Academy of Sciences of the Ukrainian SSR

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It is shown that the distribution of weakly coupled impurities over the energy levels in a relatively weak field is altered substantially if the field frequency is in one of the absorption sidebands. Depending on the relationship between the impurity, field, and phonon frequencies, the impurity can be cooled or heated strongly, and the level population can also be inverted. If the field frequency is greater than the impurity frequency, then phonon magnification occurs at the difference frequency in a definite field range. Analogously, by inducing a decay into magnons by a longitudinal or transverse field, the impurity spin distribution in ferromagnets can be changed and magnon amplification obtained. Absorption of the nonresonant radiation is saturated as the field increases.

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INTRODUCTION

In addition to the narrow phonon-free lines in the light absorption spectra of weakly coupled impurities, there are usually broad sidebands corresponding to absorption with simultaneous excitation of the impurity and phonon creation or annihilation. Actually, a new relaxation channel occurs for an impurity in a field whose ω lies in the sideband domain, i.e., decay processes in which the transition between levels is accompanied by photon and phonon generation or annihilation. If the field is sufficiently large, then the induced decays can become dominant. The impurity distribution over the energy levels on this case is determined by the frequency of the phonons participating in the decay and by the temperature and is described, as will be shown below, by the temperature $T^* = T\omega_0/(\omega_0 \pm \omega)$, where $\hbar\omega_0$ is the spacing between levels. The case when the decay process corresponds to simultaneous phonon generation and impurity excitation is special. If such decays are dominant, then the effective impurity temperature turns out to be negative $T^* = -T\omega_0/(\omega - \omega_0)$ and a population inversion occurs. Analogously, localized spin excitations interacting with magnons in ferromagnets can be cooled, heated, and inverted by a nonresonant field.

The mentioned mechanism to change the impurity temperature is similar to the cooling mechanism, for a low-frequency two-level subsystem, proposed in Ref. 1, by a field with frequency $\omega = \omega_1 - \omega_0$ ($\hbar\omega_1$ is the energy

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of a high-frequency two-level subsystem interacting with the low-frequency subsystem under consideration). As noted in Ref. 2, cooling is possible if and only if the high-frequency subsystem is rapidly relaxing (the criterion is presented below). In addition to the cooling considered in Ref. 1, a population inversion can be obtained by using a field in the case of interacting impurity subsystems with substantially distinct relaxation rates. For this the field frequency should equal the sum of the subsystem frequencies and the inversion will occur in that subsystem which relaxes more slowly.

The width of the resonant impurity absorption peaks also changes in a nonresonant field because of the induced decays, and light amplification of the frequency $T^* < 0$ occurs for ω_0. Moreover, for $\omega > \omega_0$ and $T^* > 0$ the impurity can magnify phonons (magnons) by resonance at the frequency $\omega - \omega_0$, and for $T^* < 0$ at the frequencies $\omega + \omega_0$.

1. KINETIC EQUATION AND STATIONARY DISTRIBUTION IN A MONOCHROMATIC FIELD

To analyze the influence of the induced decays on impurity dynamics, we use the simplest impurity spin model. The isolated spin Hamiltonian in a field has the form

$$H_0 = \omega_0 S_z - f(t) S_x, \quad f(t) = f_0 e^{-i\omega t} + f_0^* e^{i\omega t}; \quad \hbar = 1. \tag{1}$$

The alternating field f is nonresonant and $\omega \neq \omega_0$. If $S = 1/2$, then (1) describes an arbitrary two-level system ("energetic spin") with an electric or magnetic dipole transition between levels. For not too strong fields $|f_0| t_c \ll 1$, $t_c^{-1} = \min\{\omega, \omega_0 | \omega - \omega_0|\}$ and a slow change in the field amplitude with time $(d/dt) \ln|f_0| \ll t_c^{-1}$ the explicit time dependence of the spin operators in the Heisenberg representation can be found by using a method analogous to the method of taking the average in the theory of nonlinear vibrations,[3] by expanding $S_\varkappa(t)$ in series:

$$S_\varkappa(t) = \sum_{m,n} S_\varkappa^{(m,n)}(t) e^{i(m\omega_0 + n\omega)t}, \quad \dot{S}_\varkappa^{(m,n)} t_c \ll 1, \quad \varkappa = x, y, z. \tag{2}$$

To the first order in the field

$$\begin{aligned} S_z(t) &= \tilde{S}_z(t) + \tilde{S}_+(t)\varphi(t) + \tilde{S}_-(t)\varphi^*(t); \\ S_-(t) &= \tilde{S}_-(t) - 2\tilde{S}_z(t)\varphi(t); \quad S_+(t) = \tilde{S}_+(t) - 2\tilde{S}_z(t)\varphi^*(t); \\ \dot{\tilde{S}}_z &= 0; \quad \dot{\tilde{S}}_\pm = \pm i\omega_0 \tilde{S}_\pm; \\ S_\pm &= S_x \pm iS_y; \quad [\tilde{S}_z, \tilde{S}_\pm] = \pm \tilde{S}_\pm; \\ \varphi(t) &= \frac{1}{2}\left[\frac{f_0}{\omega_0 - \omega} e^{-i\omega t} + \frac{f_0}{\omega_0 + \omega} e^{i\omega t}\right]. \end{aligned} \tag{3}$$

The terms with the combined frequencies in (3) are responsible for the sidebands in the light absorption spectra, and result in a new important relaxation channel in comparatively strong fields. To the second order in f the frequency ω_0 is renormalized (Stark effect) $\omega_0 \to \omega_0 + 2\omega_0 |f_0|^2/(\omega_0^2 - \omega^2)$, and the commutation relations between operators change: $S_\pm$ act as the operators $S_\pm(1 - \Phi)$, while $\tilde{S}_z$ act as $S_z(1 - 2\Phi)$, where $\Phi = \frac{1}{2}|f_0|^2(\omega_0^2 + \omega^2)(\omega_0^2 - \omega^2)^{-2}$.

Moreover, terms at other combination frequencies appear in $S_\varkappa$. However, these terms are insignificant in moderate fields and for weak coupling to phonons.

The simplest Hamiltonian describing phonon relaxation has the form

$$\begin{aligned} &H = H_0 + H_{ph} + H_i, \quad H_{ph} = \sum_k \omega_k b_k^+ b_k; \\ &H_i = H_i^{(1)} + H_i^{(2)}; \quad H_i^{(1)} = \sum_k [u_k S_+ (b_k + b_k^+) + \text{h.c.}]; \\ &H_i^{(2)} = \sum_k v_k S_z (b + b_k^+), \end{aligned} \tag{4}$$

where k enumerates the vibration of the continuous spectrum in a nonideal crystal. The term $H_i^{(1)}$ specifies the transition between impurity levels with phonon participation which results in energy relaxation; in the case of deep centers this relaxation can be related to radiative decays and the analogous photon operators should be substituted in $H_i^{(1)}$ instead of the phonon operators b_k, b_k^+. The term $H_i^{(2)}$ determines the local deformation occuring during impurity excitation, and for weak coupling ($\sum_k |v_k|^2 \omega_k^{-1} \ll \omega_{ph}$, ω_{ph} is the phonon frequency essential for relaxation) in the absence of a field it has practically no influence on the energy relaxation as well as on the linewidth of impurity light absorption. Because there are terms at the frequencies $\pm\omega \pm \omega_0$ in $S_z(t)$ in (3), in the presence of a field the interaction $H_i^{(2)}$ can result in decay relaxation if one of the combined frequencies lies in the range of phonon frequencies.

The kinetic equation for the subsystem density matrix ρ in the interaction representation to the second order in H_i has the form

$$\begin{aligned} &\frac{\partial \rho}{\partial t} = -\Gamma_f \{(\bar{n}_f + 1)[\{\tilde{S}_+\tilde{S}_-, \rho\}_+ - 2\tilde{S}_-\rho\tilde{S}_+] \\ &+ \bar{n}_f [\{\tilde{S}_-\tilde{S}_+, \rho\}_+ - 2\tilde{S}_+\rho\tilde{S}_-]\} - \Gamma_\mu [\{\tilde{S}_z^2, \rho\}_+ - 2\tilde{S}_z\rho\tilde{S}_z], \\ &\tilde{S}_z \equiv \tilde{S}_z(t); \quad \{A, B\}_+ = AB + BA; \quad \Gamma_f = \Gamma + \Gamma_+ + \Gamma_- - \Gamma_{inv}; \\ &\Gamma = \pi \sum_k |u_k|^2 \delta(\omega_0 - \omega_k); \\ &\Gamma_\pm = \frac{\pi |f_0|^2}{4(\omega_0 \pm \omega)^2} \sum_k |v_k|^2 \delta(\omega_0 \pm \omega - \omega_k); \\ &\Gamma_{inv} = \frac{\pi |f_0|^2}{4(\omega - \omega_0)^2} \sum_k |v_k|^2 \delta(\omega - \omega_0 - \omega_k); \\ &\Gamma_\mu = \pi |f_0|^2 [2\bar{n}(\omega) + 1] \sum_k \left| \frac{u_k}{\omega_0 - \omega} + \frac{u_k^*}{\omega_0 + \omega} \right|^2 \delta(\omega - \omega_k); \\ &\Gamma_f t_c, \quad \Gamma_\mu t_c \ll 1; \\ &\bar{n}_f = \Gamma_f^{-1} [\Gamma \bar{n}(\omega_0) + \Gamma_+ \bar{n}(\omega_0 + \omega) + \Gamma_- \bar{n}(\omega_0 - \omega) \\ &\qquad + \Gamma_{inv}(\bar{n}(\omega - \omega_0) + 1)]; \\ &\bar{n}(\omega) = \frac{\theta(\omega)}{\exp(\omega/T) - 1}, \quad \theta(x) = \begin{cases} 0, & x < 0 \\ 1, & x \geqslant 0. \end{cases} \end{aligned} \tag{5}$$

The quadratic term in the interaction parameters, of the type $i[P_1\tilde{S}_z + P_2\tilde{S}_z^2, \rho]$ which describes renormalization of the spectrum, is not written down in (5); it does not affect the impurity energy relaxation. Despite the fact that the condition $|f_0| t_c \ll 1$ was used in deriving (5), the parameter $(\Gamma_f - \Gamma)/\Gamma$ can be arbitrary: the weakness of the field can be compensated if the phonon density of states at the combined frequencies is much greater than at the fundamental, or if $|v_k| > |u_k|$.

In a quasimonochromatic field when $\dot{\bar{n}}_f/dt \ll \Gamma_f$, Equation (5) evidently has the quasistationary solution

$$\rho_{st} = Z^{-1}\exp(-\omega_0\tilde{S}_z/T^*); \quad T^* = \omega_0\left(\ln\frac{\bar{n}_f+1}{n_f}\right)^{-1},$$
$$Z = \exp(\omega_0 S/T^*)\frac{1-\exp[-\omega_0(2S+1)/T^*]}{1-\exp(-\omega_0/T^*)}. \quad (6)$$

According to (6), the stationary distribution of a nonequilibrium impurity turns out to be a Boltzmann distribution even when decays induced by the field are taken into account. The effective temperature T* can differ considerably from the phonon temperature. For $\Gamma_f - \Gamma \gg \Gamma$ it is the case that $T^* = T\omega_0/(\omega_0 \pm \omega)$, i.e., both substantial heating and cooling of the impurity are possible. If $\Gamma_f < 0$, then the effective temperature becomes negative [for $\Gamma_f \to \pm 0$, $T^* \to \pm\infty$; equation (5) is not valid at the point $\Gamma_f = 0$, it is necessary to take account of higher orders in H_1 in the perturbation theory, and of the fact that the impurity motion is not Markovian]. It is characteristic that the population inversion takes place at once for all levels in the model used, where it will be the stronger, the smaller the Planck number of phonons at the frequency $\omega - \omega_0$.

The Boltzmann nature of the nonequilibrium quasistationary distribution (6) is essentially related (see the Appendix) to the assumption of an equal spacing between the impurity levels (phonons of the identical frequency take part in each act of induced decay). However, if the unequal spacing between levels Δ is not too great ($n_f\Delta/T \ll 1$), then (6) remains valid in the model taken for the spontaneous and induced decay transitions between adjacent levels.

The kinetic equation (5) is also applicable in the case of two-phonon interaction

$$\tilde{H}_i^{(2)} = \sum_{k_1 \neq k} v_{kk_1} S_z b_k^+ b_{k_1}. \quad (7)$$

To the second order in v_{kk}, taking account of (7) is limited by the addition of the parameters $\tilde{\Gamma}_\mu$, $\tilde{\Gamma}_\pm$, $\tilde{\Gamma}_{inv}$ to the corresponding parameters without waves:

$$\tilde{\Gamma}_\mu = \pi\sum_{kk_1}|v_{kk_1}|^2\delta(\omega_k-\omega_{k_1})\,\bar{n}(\omega_k)[\bar{n}(\omega_k)+1];$$
$$\tilde{\Gamma}_\pm = \frac{\pi|f_0|^2}{4(\omega_0\pm\omega)^2}\sum_{kk_1}|v_{kk_1}|^2\delta(\omega_0\pm\omega+\omega_{k_1}-\omega_k)[\bar{n}(\omega_{k_1})-\bar{n}(\omega_k)];$$
$$\tilde{\Gamma}_{inv} = \frac{\pi|f_0|^2}{4(\omega-\omega_0)^2}\sum_{kk_1}|v_{kk_1}|^2\delta(\omega-\omega_0+\omega_{k_1}-\omega_k)[\bar{n}(\omega_{k_1})-\bar{n}(\omega_k)]. \quad (8)$$

Let us note that modulation broadening of Γ_μ, which determines relaxation of the phase difference of the impurity wave functions, often introduces the main contribution to the half-width of the resonance absorption line for the case of high-frequency transitions $\omega_0 \gg \omega_{ph}$ at not too low temperatures.[4]

A change in the distribution because of the decays induced by the field can occur even for weakly coupled impurity spins (local or quasilocal spin excitations) in ferromagnets. The impurity spin exchange interaction with magnons ($T \ll T_C$) is described by the Hamiltonian

$$\tilde{H}_i = \tilde{H}_i^{(1)} + \tilde{H}_i^{(2)}; \quad \tilde{H}_i^{(1)} = \sum_k \tilde{u}_k S_+ b_k^+ + \text{h.c.}, \quad (4a)$$

where, in contrast to the above, b_k^+, b_k are now magnon creation and annihilation operators.[5] For a ferromagnetic impurity $\omega_0 < 0$ in (1), and an antiferromagnetic impurity $\omega_0 > 0$ in (1), three-magnon processes must be taken into account even for quasilocal excitations in order to obtain the finite width Γ. In an alternating magnetic field transverse to the static field, the dynamics of a ferromagnetic impurity is described by (5) and (8), where the operators $\tilde{S}_+$ and $\tilde{S}_-$ in (5) should be interchanged and ω_0 in (5), (8), and the expression for T* in (6) should be replaced by $|\omega_0|$. In the case of an antiferromagnetic impurity, the form of the kinetic equation agrees with (5), and (6) and (8) are applicable directly.

As is seen from (8), two-magnon decay processes induced by a transverse field freeze out with a reduction in temperature. Hence, it is expedient to use alternating magnetic fields parallel to the static field to obtain cooling or heating of the impurity spins at low temperatures. The impurity Hamiltonian then has the form

$$\tilde{H}_0 = \omega_0 S_z - f(t)S_z; \quad f(t) = f_0 e^{-i\omega t} + f_0^* e^{i\omega t}. \quad (9)$$

In a longitudinal field, the terms at the frequencies $\pm\omega \pm \omega_0$ appear in first order perturbation theory in the operators

$$S_\pm(t) \approx \tilde{S}_\pm(t)\left[1 \pm \frac{1}{\omega}(f_0 e^{-i\omega t} - f_0^* e^{i\omega t})\right].$$

These terms result in induced single-magnon decays, which are to a known degree analogous to the single-phonon decays considered above in the transverse field.[1)] The essential difference between these decay processes is due to the absence of terms of the type of S_+b_k, $S_-b_k^+$ in $\tilde{H}_i^{(1)}$ in (4a). Because of the absence of such terms, single-magnon decay processes of the type of $\omega = |\omega_0| + \omega_k$, resulting in a population inversion in sufficiently strong fields, are forbidden for ferromagnetic impurities, while on the other hand, only such processes are allowed for antiferromagnetic impurities. Hence, the population inversion of a localized antiferromagnetic spin excitation is easily obtained by using a longitudinal field.

To obtain significant cooling of a low-frequency impurity by "subconnecting" it to high-frequency phonons (magnons) by a field is difficult if ω_0 is very much smaller than the quasiparticle bandwidth since the population-inversion process [Γ_+ in (5)] occurs together with the cooling process [it is described by the parameter Γ_{inv} in (5)], and these processes mutually cancel (the inversion is forbidden by selection rules in the case of a low-frequency ferromagnetic impurity in a longitudinal field and there is no cancellation). Hence, the resonance mechanism proposed in Ref. 1, when $\omega \approx \omega_1 - \omega_0$ ($\omega_1 \gg \omega_0$) and the impurity subsystem I is in good contact with a thermostat so that its damping is $\Gamma_1 \gg \Gamma$, plays an important part in cooling. If $\Gamma_1 \ll \omega_0$, then the resonance field with frequency $\omega_1 - \omega_0$ does not result in the inversion process in practice, and $\Gamma_+ \gg \Gamma_{inv}$.

An electron subsystem and a local (quasilocal) impurity vibration are an example of interacting subsystems with substantially different relaxation times. An electron

subsystem can often be considered two-leveled and described by the Hamiltonian (1) (the "energy spin", $S = 1/2$). The Hamiltonian local vibration (LV) interacting with an electric transition has the form

$$H_L = \omega_L a^+ a + V S_z (a + a^+), \quad S = 1/2, \tag{10}$$

where a^+, a are the LV creation and annihilation operators, and V is the interaction parameter $|V| \ll \omega_L, \omega_0$. The polarizability in an electron subsystem is usually larger than in the LV, and the field term is omitted in (10). To the first order in V and the second order in the interaction with vibrations of a continuous crystal spectrum, the kinetic equation in the interaction representation has the form

$$\frac{\partial \rho}{\partial t} = -\hat{\Gamma}_s \rho - \hat{\Gamma}_L \rho - i \frac{V}{2\omega_L} [(a \tilde{S}_r f_r e^{i\Omega_r t} + \text{h.c.}), \rho]. \tag{11}$$

The operator $\Gamma_s \rho$ in (11) describes the damping of the electron subsystem and is determined by (5). The operator $\hat{\Gamma}_L \rho$ describes LV damping and can be obtained from the right side of (5) by replacement of the operators $\tilde{S}_+$ and $\tilde{S}_-$ by a^+ and a, respectively, $\bar{n}_f$ by $\bar{n}(\omega_L)$, Γ_f by Γ_L (Γ_L is the LV decay damping parameter). The last term on the right side of (11) describes the induced resonant interaction of the LV and the electron subsystem; for $\omega_L \sim \omega_0 \pm \omega$ we have $\tilde{S}_r = \tilde{S}_+$, $\Omega_r = \omega_0 - \omega_L \pm \omega$, but for $\omega \sim \omega_L + \omega_0$ we have $\tilde{S}_r = \tilde{S}_-$, $\Omega_r = \omega - \omega_L - \omega_0$; $f_r = \pm f_0^*$ for $\omega \sim \omega_L \pm \omega_0$ and $f_r = f_0$ for $\omega \sim \omega_0 - \omega_L$. The nonresonant term $iV[(\tilde{S}_z a e^{-i\omega_L t} + \text{h.c.}), \rho]$ is omitted in (11); to second order in perturbation theory it results in a shift $\sim V^2/\omega_L$ of the electron levels.

The dynamics of the electron subsystem is described by the density matrix $\rho_s = \text{Tr}_L \rho$, where Tr_L denotes the trace in the LV wave functions. According to (11) we have

$$\frac{\partial \rho_s}{\partial t} = -\hat{\Gamma}_s \rho_s - i \frac{V}{2\omega_L} [\tilde{S}_r f_r e^{i\Omega_r t}, \tag{12}$$

$$\text{Tr}_L (\rho a)] - i \frac{V}{2\omega_L} [\tilde{S}_r^+ f_r^+ e^{-i\Omega_r t}, \quad \text{Tr}_L (\rho a^+)] .$$

[Here we use the obvious equation $\text{Tr}_L(\hat{\Gamma}_L \rho) = 0$].

For $\Gamma_L \gg \Gamma$, $|V f_0/\omega_L|$ the local fluctuation "adiabatically" becomes a slowly relaxing electron subsystem. In the zero approximation according to V, the Eq. (11) has quasistationary solution (in the time scale Γ_L^{-1}):

$$\rho_0 (t) = [\bar{n}(\omega_L) + 1]^{-1} \exp(-\omega_L a^+ a / T) \rho_s (t).$$

To the first order in V equation (11) can be solved by perturbation theory by considering $\rho_s(t) \approx \text{const}$. Then

$$\text{Tr}_L (\rho a) = -i \frac{V}{\Gamma_L - i\Omega_r} \frac{f_r^*}{2\omega_L} \{[\bar{n}(\omega_L) + 1] \tilde{S}_r^+ \rho_s (t) - \bar{n}(\omega_L) \rho_s (t) \tilde{S}_r^+\}. \tag{13}$$

Substituting (13) and an analogous expression for $\text{Tr}_L \cdot (\rho a^+)$ into (12), we obtain (5) for ρ_s, where

$$|\Gamma_f - \Gamma| = \frac{V^2 |f_0|^2}{4\omega_L^2} \frac{\Gamma_L}{\Gamma_L^2 + \Omega_r^2} = \begin{cases} \Gamma_\pm, & \omega_L \sim \omega_0 \pm \omega \\ \Gamma_{\text{inv}}, & \omega \sim \omega_0 \pm \omega_L \end{cases}.$$

Therefore, the rate of the decay processes induced by a field is proportional to V^2/Γ_L in a resonance situation and is large for small Γ_L (the width of the phonon band Γ_L performs the role of $\omega_{ph} \gg \Gamma_L$ for phonon decay). Depending on the relationship between ω_0, ω_L, and ω in a slowly relaxing subsystem, considerable cooling, heating, or inversion can be obtained by using relatively weak fields.

2. PHONON, MAGNON, AND ELECTROMAGNETIC WAVE AMPLIFICATION

The change in relaxation and impurity distribution over energy levels results in a number of effects observable experimentally. Let us examine some for the simplest, but still important, case of a two-level system, $S = 1/2$. The redistribution over levels is primarily reflected in the absorption by the nonresonant field system. The power drawn from the field in the stationary mode equals

$$W = \frac{2\omega}{2\bar{n}_f + 1} \{\Gamma_+ [\bar{n}_f - \bar{n}(\omega_0 + \omega)] - \Gamma_- [\bar{n}_f - \bar{n}(\omega_0 - \omega)]$$

$$+ \Gamma_{\text{inv}} [\bar{n}_f + \bar{n}(\omega - \omega_0) + 1]\} \tag{14}$$

(which is obtained most easily by calculating the change in phonon energy because of interaction with an impurity). If just one kind of induced decays exists and one term in the braces is nonzero in (14) then, as is easily seen, saturation of the nonresonant radiation absorption, similar to the known saturation of resonance radiation absorption and having the same field dependence, will occur in strong fields ($|\Gamma_f - \Gamma| \gg \Gamma$). For instance, for $\Gamma_\pm = 0$

$$W = 2\omega \Gamma \Gamma_{\text{inv}} \frac{[\bar{n}(\omega_0) + \bar{n}(\omega - \omega_0) + 1}{\Gamma [2\bar{n}(\omega_0) + 1] + \Gamma_{\text{inv}} [2\bar{n}(\omega - \omega_0) + 1]}, \quad \Gamma_{\text{inv}} \propto |f|^2, \tag{15}$$

and the power absorbed in strong fields is independent of the field intensity:

$$W \simeq 2\omega \Gamma \frac{\bar{n}(\omega_0) + \bar{n}(\omega - \omega_0) + 1}{2\bar{n}(\omega - \omega_0) + 1}.$$

An important feature of saturation of nonresonance radiation absorption is the possibility of obtaining a population inversion of the impurity levels for $\Gamma_{\text{inv}} > \Gamma$. Pumping a two-level impurity in the sideband is quite effective (possibly more effective than pumping in a three-level scheme) since each act of light absorption transfers the impurity to an excited level which may be the working level for a laser at the resonant frequency.

If the pumping is not too strong and there is no population inversion, then for $\omega > \omega_0$ the impurity can amplify phonons (magnons) at the frequency $\omega - \omega_0$. Evaluation of the amplification factor is associated with a computation of the impurity two-time correlation functions and their spectral distributions. In a monochromatic field, a stationary mode is built up in the subsystem in the time $t \gg \Gamma_f^{-1}$, and the correlation functions ω^{-1} amplified in the period $\langle A(t)\, B(\tau) \rangle$ [denoted below by $A(t)\, B(\tau)$] depend only on the time difference $A(t)\, B(\tau) = A(t -$

τ)B(0). The correlation functions can easily be obtained by solving (5), for instance:

$$S_{zz}(t) = \overline{S_z(t)\,[S_z(0) - \bar{S}_z]} \approx \frac{\bar{n}_f(\bar{n}_f+1)}{(2\bar{n}_f+1)^2}\exp[-2\Gamma_f(2\bar{n}_f+1)t] + \frac{e^{-\gamma_f t}}{2\bar{n}_f+1}[(\bar{n}_f+1)\Phi(t)e^{-i\omega_0 t} + \bar{n}_f\Phi^*(t)e^{i\omega_0 t}];$$

$$\gamma_f = \Gamma_f(2\bar{n}_f+1) + \Gamma_\mu;$$

$$\Phi(t) = \frac{|f_0|^2}{4}[(\omega_0-\omega)^{-2}e^{i\omega t} + (\omega_0+\omega)^{-2}e^{-i\omega t}]. \qquad (16)$$

It is seen from (16) that damping of the correlation functions in the field grows because of both the induced delay processes ($\Gamma_f[2\bar{n}_f + 1] > \Gamma[1 + 2\bar{n}(\omega_0)]$) and the modulation processes (the parameter Γ_μ). The amplitudes of the correlators also change, and to the second order in the field terms at the frequencies $\pm\omega \pm \omega_0$ occur in $S_{zz}(t)$ (the small correction $\sim \Phi(0)$, compared with unity and independent of time, is omitted in the first term in (16), but the parameter $(\Gamma_f - \Gamma)/\Gamma$ is arbitrary).

The phonon absorption (amplification) induced by the field (1) at the combined frequencies is due to the adiabatic interaction $H_1^{(2)}$ in (4). The coefficient of stationary absorption of phonons of frequency Ω is determined by the expression $q(\Omega)\sum_k f_k^2 |v_k|^2 \delta(\Omega - \omega_k)$ (f_k^2 is a weight function dependent on the polarization of the waves being absorbed, their propagation directions, etc.), where

$$q(\Omega) = \mathrm{Re}\int_0^\infty dt e^{i\Omega t}\,\overline{[S_z(t),\ S_z(0)]}, \qquad (17)$$

and according to (16)

$$q(\Omega) = \frac{|f_0|^2}{4(2\bar{n}_f+1)(\omega_0\pm\omega)^2}\,\frac{\gamma_f}{\gamma_f^2 + [\Omega - (\omega_0 \pm \omega)]^2}, \quad \Omega \sim \omega_0 \pm \omega;$$
$$q(\Omega) = -\frac{|f_0|^2}{4(2\bar{n}_f+1)(\omega-\omega_0)^2}\,\frac{\gamma_f}{\gamma_f^2 + [\Omega - (\omega-\omega_0)]^2}, \quad \Omega \sim \omega - \omega_0. \qquad (18)$$

As is seen from (18), the form of the induced phonon absorption peak $q(\Omega)$ is Lorentzian with half-width γ_f, and the peak intensity is determined by the factor $(2\bar{n}_f + 1)^{-1}$. Until the inverting decays dominate and $\Gamma_f > 0$ [then $\bar{n}_f > 0$ according to (5)] the nonequilibrium impurity absorbs phonons at the frequencies $\omega_0 \pm \omega$ but coherently amplifies phonons at the difference frequency $q(\omega - \omega_0) < 0$. If the inverting processes are dominant and $\Gamma_f < 0$ (then $\bar{n}_f < -1$), phonons at the frequency $\sim \omega - \omega_0$ are then absorbed.[2)] The field dependence of the coherent amplification of phonons at the difference frequency at $\Gamma_\pm = 0$ is the following when modulation broadening is neglected:

$$-q(\omega-\omega_0) \propto \frac{\Gamma_{inv}(\Gamma - \Gamma_{inv})}{\{\Gamma[2\bar{n}(\omega_0)+1] + \Gamma_{inv}[2\bar{n}(\omega-\omega_0)+1]\}^2}, \qquad (19)$$

$\Gamma_{inv} \propto |f_0|^2$.

The amplification is a maximum for $\Gamma_{inv} = \Gamma[2\bar{n}(\omega_0) + 1]/[4n(\omega_0) + 2\bar{n}(\omega - \omega_0) + 3]$, and for $\Gamma_{inv} = \Gamma$ it is replaced by absorption which grows monotonically with the field.

The application of nonresonant brightneing to produce a phonon maser at a three-level impurity has been proposed in Ref. 6. However, the spontaneous emission of phonons was actually computed in this work only for $\Gamma_{inv} \ll \Gamma$. At the same time, estimates were made for sufficiently strong fields ($> 10^4$ V/cm) for the Cr^{3+} impurity in Al_2O_3, where Γ is small, and apparently $\Gamma_{inv} \gg \Gamma$ in such fields. Let us note that the maximum value of the phonon gain coefficient is independent of Γ for $\Gamma \gg \Gamma_\mu$. Favorable conditions for tunable coherent amplification of phonons exist in the case of pumping of low-frequency impurities ($\omega_0 \ll \omega_{ph}$), where $\Gamma_+ \sim \Gamma_{inv}$ and there is no heating (and inversion even more so). In this case the gain coefficient at the frequency $\omega - \omega_0$ is positive and increases with the field in proportion to $|f_0|^2/[1 + A|f_0|^2]$ [$A \sim (\Gamma_+ + \Gamma_{inv})/\Gamma|f_0|^2$ is independent of the field].

Absorption of light and phonons at the impurity natural frequency can be examined analogously to absorption at the combined frequencies. The form of the absorption peak is determined by the function

$$\tilde{q}(\Omega) = \mathrm{Re}\int_0^\infty dt e^{i\Omega t}\,\overline{[S_-(t), S_+(0)]};$$
$$\tilde{q}(\Omega) \simeq \frac{1}{2\bar{n}_f+1}\,\frac{\gamma_f}{\gamma_f^2 + (\Omega-\omega_0)^2}, \quad \Omega \sim \omega_0. \qquad (20)$$

The absorption peak broadens with the increase in the nonresonant field. For $\Gamma_{inv} > \Gamma$, when $\bar{n}_f < -1$ and there is a population inversion $\bar{q}(\Omega) < 0$, i.e., absorption is replaced by amplification. Other conditions being equal, the maximum gain is achieved at low temperatures $\bar{n}(\omega - \omega_0) \ll 1$, when the inversion is maximal and γ_f is minimal.

The form of the peak of magnon absorption by local or quasilocal spin excitations in ferromagnets is determined by the following function when exchange interaction is taken into account

$$q_m(\Omega) = \mathrm{Re}\int_0^\infty dt e^{i\Omega t}\,\overline{[S_+(t),\ S_-(0)]} \qquad (21)$$

[the absorption coefficient is quadratic in the parameter $\bar{u}_k$ in (4a)]. As is seen from (3), magnon absorption or amplifications can occur only at the resonant frequency and only by a ferromagnetic impurity ($\omega_0 < 0$) in a transverse external field, where a population inversion is necessary for amplification. For $S = 1/2$ the expression for $q_m(\Omega)$ for a ferromagnetic impurity agrees with (20) (ω_0 must be replaced by $|\omega_0|$). If the pumping is by a longitudinal field and the Hamiltonian has the form (8), then in addition to absorption of the natural frequency, absorption at the combined frequencies is also possible. Since the longitudinal field with induced three-magnon processes neglected does not result in a population inversion of the ferromagnetic impurity levels, then there is no gain to the second order in the field. An antiferromagnetic impurity can amplify magnons at the frequency $\omega - \omega_0$ if the alternating magnetic field is not too large and there is no inversion

$$q_m(\Omega) = -\frac{1}{2\bar{n}_f+1}\,\frac{|f_0|^2}{\omega^2}\,\frac{\gamma_f}{\gamma_f^2 + [\Omega - (\omega-\omega_0)]^2}, \qquad (22)$$
$\Omega \sim \omega - \omega_0, \quad S = 1/2.$

The field dependence of such amplification is analogous to that considered above for phonon amplification.

A change in the impurity spin distribution in ferromagnets and in their relaxation parameters can be recorded by means of the change in both the form and width of their resonance absorption spectra, and the width of the Mössbauer lines at the impurity nuclei. As is shown in Ref. 7, the Mössbauer linewidth is proportional to the spectral distribution of the correlation function $S_{zz}(t)$ at zero frequency. According to (16)

$$\int_0^\infty dt S_{zz}(t) \approx \frac{\bar{n}_f(\bar{n}_f+1)}{2\Gamma_f(2\bar{n}_f+1)^3}$$

depends strongly on the nonresonant field amplitude and its frequency, where both narrowing and broadening of the Mössbauer line are possible. Let us note that $f_0/(\omega-\omega_0)$ enters into the expression for the induced relaxation rate when pumping localized spins by a transverse field, while f_0/ω enters for pumping by a longitudinal field. If $\omega_0 \gg \omega_{mag}$, then $(\omega_0-\omega)^{-1} \sim \omega_{mag}^{-1} \gg \omega^{-1} \sim \omega_0^{-1}$, and pumping by a transverse field can be more effective.

CONCLUSION

Impurity excitation by a nonresonant field generally requires a greater field intensity than for resonance pumping: If $|\Gamma_f - \Gamma| \sim \Gamma$, then $f_0/f_{res} \sim \sqrt{\omega\varkappa(\omega)/\omega_0\varkappa(\omega)}$, where f_{res} is the resonant field intensity resulting in saturation of the absorption; $\varkappa(\omega)$ is the impurity absorption coefficent measured experimentally [for $(\omega-\omega_0) \gg \Gamma$ the cross terms in the impurity and the phonons in the system polarizability due to the addition of $f(t)S_\varkappa\sum_k m_k(b_k+b_k^+)$ to the system Hamiltonian in the external field,[3] introduce a contribution to impurity absorption, where taking account of this addition reduces to renormalizing Γ_f]. However, nonresonant, compared to resonance, pumping has three advantages in every case. First, it permits obtaining a population inversion, where interacting subsystems with substantially different but sufficiently large relaxation times, for instance, the high-frequency electron transition and local vibration or nuclear and electronic spin times, are especially convenient to obtain a population inversion. Second, both incoherent (luminescence type) and coherent tunable generation of high-frequency phonons or magnons are possible in a nonresonant field. It is expedient to use either low-frequency impurities ($\omega_0 < \omega_{ph}$) so that one "useless" phonon at the frequency ω_0 would be emitted per "useful" phonon at the frequency $\omega-\omega_0$, or high-frequency impurities, where spontaneous energy relaxation is due to radiation decays. For coherent generation it is necessary to select the field intensity carefully in order to obtain the optimal mode in which phonon amplification is independent [see (18) and (19)] of either the electron-phonon coupling, or the relaxation time Γ^{-1} [if the modulation or inhomogeneous broadenings noticeably exceed Γ, then as is seen from (18), the maximum value of the gain coefficient is proportional to Γ and is achieved in weaker and weaker fields, the stronger the coupling to the phonons]. Third, nonlinear effects can be observed in a nonresonant field at greater impurity concentrations than in a resonant field since inhomogeneous broadening should not noticeably exceed γ_f in the resonance case, and ω_{ph} in the nonresonance case.

Nonresonant brightening can be used also to produce sensitive tunable phonon detectors. If the temperature is sufficiently low ($T \ll \omega_0-\omega$) and $\omega_0 \gg \omega_{ph}$, so that impurity energy relaxation is due to radiative decays, then phonons at the frequency $\omega_0-\omega$ can be detected by luminescence at the natural frequency ω_0.

APPENDIX

It is interesting to examine the dynamics of a multilevel impurity in a nonresonant field if the spacing between the levels differs substantially. The Hamiltonian of an isolated impurity in the field has the form

$$H_0 = \sum_{m>1}\omega_m a_m^+ a_m - \sum_{m+n}(f_{mn}e^{-i\omega t} + f_{mn}^* e^{i\omega t})a_m^+ a_n; \quad m, n = 0, 1, \ldots;$$
$$[a_m, a_n^+] = \delta_{mn}; \quad f_{mn} = f_{nm}; \quad \omega_m \neq \omega_n; \quad \hbar = 1, \tag{A.1}$$

where a_m^+, a_m are impurity generation and annihilation operators in the state m, and f_{mn} is proportional to the dipole moment of the transition between the states m and n.

To the first order in the field in the Heisenberg representation

$$a_m(t) = \tilde{a}_m - \sum_{n+m}\left[\frac{f_{mn}e^{-i\omega t}}{\omega_n-\omega_m+\omega} + \frac{f_{mn}^* e^{i\omega t}}{\omega_n-\omega_m-\omega}\right]\tilde{a}_n, \quad \tilde{a}_n \equiv \tilde{a}_n(t);$$
$$\dot{\tilde{a}}_m = -i\tilde{\omega}_m\tilde{a}_m; \quad \tilde{\omega}_m = \omega_m + 2\sum_{n+m}\frac{|f_{mn}|^2(\omega_m-\omega_n)}{(\omega_{mn}-\omega_n)^2-\omega^2}; \tag{A.2}$$
$$[\tilde{a}_m(t), \tilde{a}_n^+(t)] = \delta_{mn}\left\{1 - 2\sum_{p+m}|f_{mp}|^2\frac{(\omega_p-\omega_m)^2+\omega^2}{(\omega_p-\omega_m)^2-\omega^2}\right\}$$

(here second order corrections are taken into account partially in the expression for the frequency shift $\tilde{\omega}_m - \omega_m$ and the change in commutation conditions). If at least one of the frequencies $|\omega_m \pm \omega|$ falls into the phonon frequency range, then the field can induce decay into phonons of the corresponding frequency.

$$H = H_0 + H_{ph} + H_i^{(1)} + H_i^{(2)}; \quad H_{ph} = \sum_k \omega_k b_k^+ b_k;$$
$$H_i^{(1)} = \sum_{m+n} a_m^+ a_n U_{mn}(b_k, b_k^+); \quad H_i^{(2)} = \sum_{m,k} V_{mk} a_m^+ a_m (b_k + b_k^+), \tag{A.3}$$

where $H_i^{(1)}$ describes the spontaneous decay processes in the system, and $H_i^{(2)}$ is the adiabatic interaction which results, in the absence of a field, in a small shift of the weakly coupled impurity level, and to the appearance of impurity absorption sidebands.

Only the terms $\sim |V_{mk}|^2|f|^2$ will be taken into account below in examining the effect of the field on relaxation processes, and terms quadratic in f, and in the parameters $H_i^{(1)}$ will be omitted. As is seen in the spin system example, the latter do not alter the results in principle, but the parameters V_{mk} can significantly exceed the corresponding parameters $H_i^{(1)}$ in every case for a high-frequency impurity. In this approximation the kinetic equation to the second order in the interaction has the form

$$\frac{\partial\rho}{\partial t} = -\sum_{m+n}\theta(\omega_m-\omega_n)\Gamma_{mn}\{(\nu_{mn}+1)[\{\tilde{a}_m^+\tilde{a}_m, \rho\}_+ - 2\tilde{a}_n^+\tilde{a}_m\rho\tilde{a}_m^+\tilde{a}_n]$$
$$+ \nu_{mn}[\{\tilde{a}_n^+\tilde{a}_n, \rho\}_+ - 2\tilde{a}_m^+\tilde{a}_n\rho\tilde{a}_n^+\tilde{a}_m]\}; \quad \theta(x) = \begin{cases}0, & x<0\\ 1, & x\geqslant 0,\end{cases}$$

$$\Gamma_{mn} = \gamma_{mn} + \Gamma^{+}_{mn} + \Gamma^{-}_{mn} - \Gamma^{i}_{mn};$$

$$\Gamma^{\pm}_{mn} = \frac{\pi |f_{mn}|^2 \theta(\omega_m - \omega_n)}{(\omega_m - \omega_n \pm \omega)^2} \sum_k |V_{mk} - V_{nk}|^2 \delta(\omega_m - \omega_n \pm \omega - \omega_k);$$

$$\Gamma^{i}_{mn} = \frac{\pi |f_{mn}|^2 \theta(\omega_m - \omega_n)}{(\omega_m - \omega_n - \omega)^2} \sum_k |V_{mk} - V_{nk}|^2 \delta(\omega_m - \omega_n + \omega_k - \omega);$$

$$\nu_{mn} = \Gamma^{-1}_{mn} [\gamma_{mn} \bar{n}(\omega_m - \omega_n) + \Gamma^{+}_{mn} \bar{n}(\omega_m - \omega_n + \omega) + \Gamma^{-}_{mn} \bar{n}(\omega_m - \omega_n - \omega) + \Gamma^{i}_{mn} (\bar{n}(\omega - \omega_m + \omega_n) + 1)]. \quad (A.4)$$

in which γ_{mn} is the damping due to spontaneous decays, and renormalization of the frequencies ω_m due to interaction is considered performed. It is seen from (A.4) that taking account of induced decay processes does not alter the form of the kinetic equation, however, the relaxation rate can be changed considerably even in moderate fields.

According to (A.4), only the diagonal elements of the density matrix ρ differ from zero in the stationary mode, but they cannot successfully be calculated explicitly in the general case and the distribution turns out not to be a Boltzmann distribution. However, if both the spontaneous and the induced decays are related to the transition only between adjacent impurity levels, then $\rho_{m+1,\,m+1}/\rho_{m,m} = \nu_{m+1,m}/(\nu_{m+1,m} + 1)$ in the stationary mode. For $\Gamma^{i}_{m+1,m} > \gamma_{m+1,m}$ this ratio is greater than unity, i.e., a population inversion holds for this pair of levels.

NOTATION

Here, H is the Hamiltonian; ω_0 is the impurity frequency; f is the nonresonance field strength; ω is the fil field frequency; ρ is the density matrix; Γ_f is the effective decay broadening taking field-induced decays into account; T* is the effective impurity temperature; Γ_μ is the modulation broadening; ω_L is the localized vibration frequency; V is the electron-localized vibration interaction parameter; W is the radiation power absorbed by the impurity; $q(\Omega)$ is phonon absorption; $\tilde{q}(\Omega)$ is resonance radiation absorption; and $q_m(\Omega)$ is magnon absorption.

[1)]Decays of impurity paramagnetic ions into phonons in nonmagnetic crystals can also be induced by using the longitudinal field (9). The dynamics of an impurity spin in a longitudinal field is described by (5), where v_k in the expressions for $\Gamma_\pm$ should be replaced by $2u_k(\omega_0 \pm \omega)/\perp$.
[2)]Each act of induced decay described by the parameter Γ_{inv} is related to phonon generation, where the frequency of the phonons being emitted is in a band of width $\sim \gamma_f$ around the frequency $\omega - \omega_0$. The function $q(\Omega)$ defines a linear reaction of the system to the coherent flux of monochromatic phonons. The linear theory is applicable as long as the effective force acting on the impurity from these phonons, which is expressed in frequency units, is small compared to Γ_f. For $\Gamma_f < 0$ such a weak coherent phonon flux is absorbed although the impurity as a whole radiates phonons (incoherent) in the neighborhood of the frequency $\omega - \omega_0$.
[3)]Such an addition is especially important if direct electric dipole transitions are forbidden in the impurity.

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