

# Some features of the excitation spectrum of an anisotropic ferromagnetic metal in a magnetic field

V. V. Val'kov

*Kirenskiĭ Physics Institute, Siberian Division, USSR Academy of Sciences*

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The determination of the magnetic-excitation spectrum of metallic  $s$ - $f$  ferromagnets is considered with single-ion anisotropy taken exactly into account. The diagram technique for Hubbard operators is used to obtain a dispersion equation at arbitrary magnetic-field orientations. The spectrum of elementary excitations of easy-plane, easy-axis, and cubic metallic ferromagnets is calculated, in the long-wave region and in collinear geometry, for three possible orientations of the easy-magnetization axis. The gap in the spectrum of the lower branch of the transverse oscillations is determined with account taken of quantum corrections. It is shown that in a sufficiently strong external magnetic field or for sufficiently strong single-ion anisotropy, the dependence of the lower-branch excitation spectra of all the considered ferromagnet metals on the quasimomentum becomes anomalous in that the frequency decreases when the quasimomentum increases. An important factor is that the energy scale of such an anisotropy or of the magnetic field is smaller by a factor  $(\mu/4\Delta)^2$  (where  $\mu/\Delta \gg 1$ ) than the separation  $\Delta$  of the spin subbands, so that observation of the predicted anomaly is quite realistic. The onset of the anomaly and the form of the spectrum are analyzed in detail, in the whole region of the Brillouin zone, by numerically solving the dispersion equation for the isotropic limit. The conditions for the limits of the applicability of the Heisenberg model to the description of the spectrum of the magnetic excitations of a metallic ferromagnet are obtained. When these conditions are not met, effective exchange interaction between the magnetoactive ions becomes dependent both on the external magnetic field and on the anisotropy.

## 1. INTRODUCTION

In several known classes of metallic magnets the characteristic single-ion anisotropy (SA) is comparable in magnitude with the energy of exchange interaction between the magnetically active ions. Examples are the intermetallides  $\text{ReAl}_2$  (Re is a rare-earth ion),<sup>1,2</sup> and a number of actinide compounds.<sup>3</sup>

Description of such metallic magnets requires, besides a consistent allowance for the conduction electron, also consideration of the strong single-ion correlations; this hinders substantially the development of the theory, especially for SA of cubic symmetry. The spectrum of the magnetic excitations of strongly anisotropic metallic ferromagnets is therefore frequently studied under the assumption that the interactions can be considered stage by stage. Thus, during the first stage, without allowance for the external magnetic field  $H$  and the SA, an indirect exchange interaction takes place via the conduction electrons. This interaction between the rare-earth ions is described by a Heisenberg Hamiltonian (to which terms describing higher-multipole interactions are sometimes added), after which a theoretical calculation is performed with account taken of the SA and of the external magnetic field.

In fact, the effective interaction produced between RE ions in second-order perturbation theory<sup>5,6</sup> reduces strictly speaking to a Heisenberg interaction only in the absence of a magnetic field  $H$  and of SA. It is therefore not clear beforehand to what value of  $H$  or to what magnitude of the SA can the magnetic-excitation spectrum of metallic anisotropic magnets be calculated within the framework of the effective-exchange model, followed by allowance for single-ion anisotropy. There should obviously exist a parameter range in

which the effective exchange interaction becomes dependent on the anisotropy and on the field  $H$ . In this region the role of conduction becomes substantial

It was demonstrated earlier by Dzyalochinskiĭ that conduction electrons can play a nontrivial role in the magnetic properties of RE metals. He has shown that the topological singularities of the Fermi surface exert a substantial influence on the character of the magnetic ordering and determine the period of an antiferromagnetic-metal structure. When a sufficiently strong external magnetic field is applied and a transition to the ferromagnetic state takes place, the spin-wave spectrum is modified both by the direct influence of the external magnetic field on the RE ion through Zeeman interaction, and by a change of the effective interaction itself.

One can similarly expect the modification of the magnetic-excitation spectrum to become modified at high SA values via two channels. First, the presence of SA makes the structure of the single-ion levels nonequidistant. Second, under strong SA the effective exchange can become dependent on the anisotropy constant. When the anisotropy is increased one can therefore expect, besides an increase of the activation energy, also a change of the spin-wave rigidity if the anisotropy has become large enough.

The spin-wave spectrum of metallic ferromagnets can be analyzed without resorting to concept of an effective exchange interaction. In the framework of the  $s$ - $f$ ( $d$ ) model, such a theory is well known.<sup>5,6,8</sup> The  $s$ - $f$  model for the simplest case of uniaxial anisotropy and collinear geometry was considered in Ref. 9.

The present paper contains a derivation of a dispersion equation for the elementary-excitation spectrum of  $s$ - $f$  ferro-

magnets, with exact allowance for single-ion anisotropy of arbitrary symmetry. The long-wave excitation spectra of easy-plane, easy-axis, and cubic metallic ferromagnets are calculated analytically. It is shown that in a strong (but not quantizing) magnetic field, or at relatively high anisotropy, an anomaly not describable in terms of the effective exchange interaction is produced in the spectrum of the lower branch of the transverse oscillations. The Heisenberg model is then invalid in principle, since the indirect interaction between the RE ions becomes dependent on the external magnetic field and on the anisotropy constants. It is important that when the anomaly arises the energy scale of the external magnetic field or of the SA is  $(\mu/4\Delta)^2$  times smaller than the separation  $\Delta$  between the spin subbands ( $\mu$  is the chemical potential of the system, with  $\mu/\Delta \gg 1$ ). The predicted anomalies of the spectral properties can therefore be experimentally investigated in many metallic ferromagnets with low Curie temperatures.

## 2. HAMILTONIAN OF AN ANISOTROPIC $s$ - $f$ MAGNET IN THE ATOMIC REPRESENTATION

Consider a rare-earth metallic magnet in which the magnetoactive  $f$  ions interact with the conduction electrons. This interaction produces an indirect exchange coupling between the localized magnetic moments of the rare-earth ions; we shall describe part of this interaction within the framework of the standard  $s$ - $f$  ( $d$ ) model.<sup>5</sup> Only magnetoactive-ion states corresponding to the lower multiplet will be considered. The influence of the crystal field in which the  $f$  ions are located will be taken into account in standard fashion, using the Wigner-Eckart theorem and the Stevens operator technique.<sup>10</sup> The Hamiltonian of the considered anisotropic  $s$ - $f$  magnet in an external magnetic field  $\mathbf{H}_0$  can then be written in the form

$$\begin{aligned} \mathcal{H} = & \sum_{\mathbf{k}\sigma} (\epsilon_{\mathbf{k}} - \mu) a_{\mathbf{k}\sigma}^+ a_{\mathbf{k}\sigma} - \sum_f (2\mu_B \mathbf{H}_0 \mathbf{s}_f + g\mu_B \mathbf{H}_0 \mathbf{J}_f) \\ & + \sum_f \sum_{nm} B_{nm} O_n^m(f) - \sum_{fg} 2A_{fg} (\mathbf{s}_f \mathbf{J}_g). \end{aligned} \quad (1)$$

The zero subscript of the magnetic-field vector means that in the initial coordinate frame connected with the crystallographic axes the components of these fields are given by

$$H_0^x = H \sin \theta_H \cos \varphi_H, \quad H_0^y = H \sin \theta_H \sin \varphi_H, \quad H_0^z = H \cos \theta_H, \quad (2)$$

where  $\theta_H$  and  $\varphi_H$  are respectively the polar and azimuthal angles that determine the orientation of  $\mathbf{H}_0$ ,  $\mathbf{J}_f$  is the angular-momentum operator of the magnetoactive ion at site  $f$ , and  $\mathbf{s}_f$  is the spin operator of the collectivized electron in the Wannier representation. The third term in (1) describes the influence of the crystal field, and  $O_n^m(f)$  are the Stevens operators.<sup>10</sup> The set of nonzero coefficients  $B_{nm}$  is determined by the crystal-field symmetry. For practical purposes it is convenient to develop a general formalism without specifying beforehand the concrete form of the single-ion anisotropy, and analyze individual cases during the final stages of the calculation. The last term of the Hamiltonian describes  $s$ - $f$  exchange coupling of itinerant and localized electrons<sup>5</sup>;  $A_{fg}$  is the integral of the exchange interaction between an itinerant electron located in the  $f$  cell (Wannier

representation) and the localized moment of an  $4f$ -shell RE ion located at site  $g$ .

Since the total  $z$  component of the total-angular-momentum operator does not commute with the Hamiltonian, the transitions between the single-ion states constructed with account taken of the SA, and the changes of the spin projections of the itinerant electrons are not connected with the angular-momentum-projection conservation law, as is the case in anisotropic<sup>5,8,11</sup> and easy-axis<sup>9</sup> ferromagnets. As a result, the number of terms increases substantially in each order of perturbation theory. Therefore the use of a diagram technique and of a fixed spin projection for the electron functions, as in Refs. 8, 9, 11 and 12, would hinder significantly an analysis of the diagram series.

The diagram series for the considered model can be greatly simplified by using matrix (in the spin indices) electron Green's functions (in analogy with the method used in superconductivity theory<sup>13</sup>) as well as the method developed in Refs. 14 and 15 to take into account an SA of arbitrary symmetry in the theory of Heisenberg magnets.

We change to new coordinates in which the localized-subsystem magnetic moment is oriented along the  $z$  axis. This is effected by the unitary transformation

$$\mathcal{H} \rightarrow \mathcal{H}' = U \mathcal{H} U^+, \quad (3)$$

where

$$U = \prod_f \exp(i\theta T_f^y) \exp(i\varphi T_f^z), \quad \mathbf{T}_f = \mathbf{J}_f + \mathbf{s}_f.$$

The azimuthal angle  $\varphi$  and the polar angle  $\theta$  describe the direction of the vector  $\langle \mathbf{J}_f \rangle$  in the initial coordinate frame. Under this transformation the Hamiltonian (1) retains its preceding form except that  $\mathbf{H}_0$  must be replaced by the vector  $\mathbf{H}$ :

$$H^x = H [\cos \theta \sin \theta_H \cos (\varphi_H - \varphi) - \cos \theta_H \sin \theta],$$

$$H^y = H \sin \theta_H \sin (\varphi_H - \varphi),$$

$$H^z = H [\sin \theta \sin \theta_H \cos (\varphi_H - \varphi) + \cos \theta_H \cos \theta], \quad (4)$$

and the operators  $O_n^m(f)$  by

$$\tilde{O}_n^m(f) = U O_n^m(f) U^+. \quad (5)$$

The Hamiltonian of the itinerant electrons, with allowance for the external magnetic field and for the additional magnetization by the  $s$ - $f$  ( $d$ ) exchange interaction, is in general not diagonal in the spin indices. The physical reason is that in an arbitrary geometry the effective field acting on the itinerant electrons is not directed along the new  $z$  axis. Obviously, the change to the proper quantization axis of the collectivized electrons results in diagonalization in the spin indices. This change can be effected by using the known  $u$ - $v$  transformation:

$$a_{\mathbf{k}\uparrow} = u c_{\mathbf{k}\uparrow} - v c_{\mathbf{k}\downarrow}, \quad a_{\mathbf{k}\downarrow} = v^* c_{\mathbf{k}\uparrow} + u c_{\mathbf{k}\downarrow}. \quad (6)$$

The collectivized-electron Hamiltonian takes then the diagonal form

$$\mathcal{H}_0 = \sum_{\mathbf{k}\sigma} (\epsilon_{\mathbf{k}} - \mu - \sigma \Delta) c_{\mathbf{k}\sigma}^+ c_{\mathbf{k}\sigma}, \quad (7)$$

if the transformation parameters (6) are given by

$$u = [(1+x)/2]^{1/2}, \quad v = \left( \frac{H_-}{H_\perp} \right) [(1-x)/2]^{1/2},$$

$$x = H_\parallel / (H_c^2 + H_\perp^2)^{1/2},$$

$$H_c = H_x + \left( \frac{A_0}{\mu_B} \right) R_L, \quad R_L = \langle J_f^z \rangle, \quad A_0 = \sum_g A_{fg},$$

$$H^\pm = H_x \pm iH_y, \quad H_\perp^2 = H_x^2 + H_y^2, \quad \Delta = 2\mu_B (H_c^2 + H_\perp^2)^{1/2}. \quad (8)$$

For an exact allowance for the SA effect, we change to the atomic representation, in which account can be taken of all the one-ion correlations. We introduce for this purpose the one-ion states  $|\Psi_n(f)\rangle$ , which are solutions of the Schrödinger equation

$$\mathcal{H}_0(f) |\Psi_n(f)\rangle = E_n |\Psi_n(f)\rangle, \quad \mathcal{H}_0(f) = \sum_{nm} B_{nm} \sigma_n^m(f) - \bar{\mathbf{H}} \mathbf{J}_f, \quad (9)$$

where the effective field is given by

$$\bar{\mathbf{H}} = g\mu_B \mathbf{H} + 2A_0 \langle \mathbf{s} \rangle, \quad \langle \mathbf{s} \rangle = R_c \frac{(H_x, H_y, H_z)}{(H_\perp^2 + H_c^2)^{1/2}},$$

$$R_c = \frac{1}{N} \sum_{\mathbf{k}\sigma} \sigma \langle c_{\mathbf{k}\sigma}^+ c_{\mathbf{k}\sigma} \rangle.$$

Introducing in the usual manner the Hubbard operators  $X_f^{nm} \equiv |\Psi_n(f)\rangle \langle \Psi_m(f)|$ , we realize the following representation of the operators of the angular momentum  $\mathbf{J}_f$  (Refs. 16 and 17):

$$J_f^+ = \sum_\lambda \gamma_\perp(\lambda) X_f^\lambda, \quad J_f^z = \sum_\lambda \gamma_\parallel(\lambda) X_f^\lambda, \quad (10)$$

where the index  $\lambda$  "runs through" values from the set of the root vectors  $\alpha(n, m)$  and indices  $n$  of the one-ion states.<sup>14,15</sup> If  $\lambda$  is a root vector of  $\alpha(p, q)$ , then  $X_f^\lambda \equiv X_f^{pq} (p \neq q)$ , but if  $\lambda$  corresponds to the  $n$ th ion state, then  $X_f^\lambda \equiv h_{fn} \equiv X_f^{nn}$ . The parameters of the representations are matrix elements of the angular-momentum operator in the basis of the one-ion states  $|\Psi_n\rangle$ :

$$\gamma_\perp(\lambda) = \begin{cases} \langle \Psi_n | J^+ | \Psi_m \rangle, & \lambda = \alpha(n, m) \\ \langle \Psi_p | J^+ | \Psi_p \rangle, & \lambda = p \quad (p=1, 2, \dots, 2J+1) \end{cases}$$

$$\gamma_\parallel(\lambda) = \begin{cases} \langle \Psi_n | J^z | \Psi_m \rangle, & \lambda = \alpha(n, m) \\ \langle \Psi_p | J^z | \Psi_p \rangle, & \lambda = p \quad (p=1, 2, \dots, 2J+1) \end{cases}$$

Introduction of the two-component second-quantization operators

$$c_j^\pm = (c_{j\uparrow}^\pm, c_{j\downarrow}^\pm), \quad c_j = (c_{j\uparrow}, c_{j\downarrow}),$$

and the use of the representation (10) allow us to write the Hamiltonian (1) in a form convenient for further analysis:

$$\mathcal{H} = 2A_0 N R_L R_c \frac{H_c}{(H_c^2 + H_\perp^2)^{1/2}} + \sum_{\mathbf{k}\sigma} (\varepsilon_{\mathbf{k}\sigma} - \mu) c_{\mathbf{k}\sigma}^+ c_{\mathbf{k}\sigma}$$

$$+ \sum_f \sum_n E_n h_{fn}$$

$$- \sum_{fg} \sum_\lambda \overline{\{c_f^+, \hat{\Gamma}_{fg}(\lambda) c_f\} X_g^\lambda}, \quad (11)$$

where  $\varepsilon_{\mathbf{k}\sigma} = \varepsilon \mathbf{k} - \sigma \Delta$ , and the two-row matrix  $\hat{\Gamma}_{fg}(\lambda)$  is defined as

$$\hat{\Gamma}_{fg}(\lambda) = A_{fg} [\gamma_\perp^*(-\lambda) \hat{\Gamma}_\perp + \gamma_\perp(\lambda) \hat{\Gamma}_\perp^+ + \gamma_\parallel(\lambda) \hat{\Gamma}_\parallel], \quad (12)$$

with the matrices  $\Gamma_{\perp, \parallel}$  dependent on the  $u-v$  transformation parameters:

$$\hat{\Gamma}_\perp = \begin{pmatrix} uv^*, & u^2 \\ -v^{*2}, & -v^*u \end{pmatrix}, \quad \hat{\Gamma}_\parallel = \begin{pmatrix} u^2 - |v|^2, & -uv \\ -uv^*, & |v|^2 - u \end{pmatrix}. \quad (13)$$

In the construction of the single-ion states and in the diagonalization of the itinerant-electron energy operator we took into account the effects of the self-consistent field. We have therefore retained in the term describing the interaction between the itinerant and localized electrons only the correlation part of the  $s-f$  interaction, a fact indicated by the overbar on the last term of the Hamiltonian (11). The influence of the  $s-f$  exchange correlation effects can be accounted for by perturbation theory, owing to the presence of the small parameter  $AJ/\mu \ll 1$ . In the diagram interpretation of the perturbation-theory series, inclusion of the self-consistent field in the zeroth Hamiltonian corresponds to summation of the so-called one-point diagrams.<sup>8</sup> Since the mean field was taken into account by us from the very outset, the one-point diagrams of the series will be ignored in the analysis of the diagram series. The remaining diagrams are estimated in order of magnitude, and we sum in the series mainly the terms that have no smallness of order  $AJ/\mu$  (the summation of the electron loops will be discussed below).

In collinear geometry, where there is no need for the  $u-v$  transformation (6), the matrix  $\hat{\Gamma}_{fg}(\lambda)$  takes the simpler form

$$\hat{\Gamma}_{fg}(\lambda) = A_{fg} \begin{pmatrix} \gamma_\parallel(\lambda), & \gamma_\perp^*(-\lambda) \\ \gamma_\perp(\lambda), & -\gamma_\parallel(\lambda) \end{pmatrix}. \quad (14)$$

### 3. GREEN'S FUNCTION. DISPERSION EQUATION

To investigate the spectral characteristics of an anisotropic  $s-f$  magnet, we use the method of temperature Green's functions,<sup>8</sup> in analogy with the procedure used in the isotropic case.<sup>8</sup> We introduce the Green's function of the Hubbard operators<sup>16,17</sup>:

$$D_{\alpha\beta}(f\tau; g\tau') = -\langle T_\tau \bar{X}_f^\alpha(\tau) X_g^\beta(\tau') \rangle$$

$$= \frac{T}{N} \sum_{\mathbf{k}} \exp\{i\mathbf{k}(\mathbf{R}_f - \mathbf{R}_g) - i\omega_n(\tau - \tau')\} D_{\alpha\beta}(k), \quad k = (\mathbf{k}, \omega_n). \quad (15)$$

In graphic form, the equation for the function  $D_{\alpha\beta}(k)$  is

$$\overleftrightarrow{\text{---}} = \overrightarrow{\text{---}} + \overrightarrow{\text{---}} \circlearrowleft M \overleftrightarrow{\text{---}}, \quad (16)$$

where, following Refs. 19 and 20, we have introduced the force operator in addition to the customarily employed concept of mass operator. A thick line with an arrow corresponds to the function  $D_{\alpha\beta}^0(k)$  defined by the equation

$$\overrightarrow{\text{---}} = \overrightarrow{\text{---}} + \overrightarrow{\text{---}} \circlearrowright \overrightarrow{\text{---}} \quad (17)$$

The thin line with the arrow corresponds here to the "bare" function  $D_\alpha(\omega_n) = (i\omega_n + \alpha E)^{-1}$  (see Refs. 16 and 17 for

details), and lines without arrows correspond to matrix Green's functions for the conduction electrons:

$$\hat{G}(p) = \begin{pmatrix} G_{\uparrow}(p), & 0 \\ 0, & G_{\downarrow}(p) \end{pmatrix}, \quad G_{\sigma}(p) = [i\omega_p - \varepsilon_{\sigma}(p) + \mu]^{-1}. \quad (18)$$

The dark circle in (17) corresponds to the "bare" vertex  $\hat{\Gamma}_{\mathbf{k}}(\alpha)$ , which is also the matrix (12):

$$\hat{\Gamma}_{\mathbf{k}}(\lambda) = \frac{1}{N} \sum_{\mathbf{k}} \hat{\Gamma}_{\mathbf{k}}(\lambda) \exp\{i\mathbf{k}(\mathbf{R}_r - \mathbf{R}_l)\}, \quad (19)$$

and the vertices  $\Gamma_{\mathbf{k}}(n)$  will be denoted here by light circles. The matrix formulation not only decreases the number of diagrams, but also simplifies the notation for them. Thus, in the standard approach, it would be necessary to draw in Eq. (17) four diagrams with electron loops instead of the one used by us. It must be borne in mind here that in the matrix formalism closed loops must correspond to the operation of taking the trace, over the spin indices, of the product of the elements making up the loop (see below).

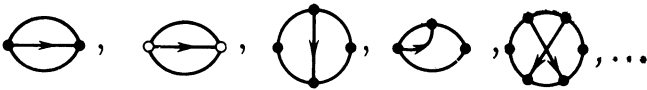
Let us write down Eq. (16) in analytic form:

$$D_{\alpha\beta}(k) = D_{\alpha\beta}^{(0)}(k) B(\beta) - \sum_{\beta_1, \beta_2} D_{\alpha\beta_1}^{(0)} M_{\beta_1\beta_2} D_{\beta_2\beta}. \quad (20)$$

If we regard  $D_{\alpha\beta}$  as components of a matrix  $D$  (Refs. 16 and 17) and define similarly the matrices  $D^{(0)}$  and  $M$ , we readily obtain from (10) the connection between the total Green's function and the mass operator:

$$D = [D^{(0)-1} + M]^{-1} B, \quad B_{\alpha\beta} = \delta_{\alpha\beta} B(\beta). \quad (21)$$

In the calculation of the Green's function  $D_{\alpha\beta}(k)$  we confine ourselves to the low-temperature region  $T \ll T_c$  ( $T_c$  is the Curie temperature). We can therefore disregard in the diagram series for the mass operator  $M$  the diagrams containing ovals, since their contribution,  $\sim \exp(-T_c/T)$ , is exponentially small.<sup>8</sup> The diagrams for  $M$ , of lower order in the interaction, take then the form



The absence of an angular-momentum-projection conservation law at the vertices causes the analytic expressions for these diagrams to contain summations over the spin indices. Except for this circumstance, the structure of the analytic expressions is similar to that for the case of an easy axis ferromagnet (FM).<sup>9</sup> For order-of-magnitude estimates of the above diagrams we can therefore use the results of Refs. 9 and 11. Thus, the first two diagrams yield a contribution  $\sim (A/\mu) \ln(\mu/A) \ll 1$  and can therefore be neglected if it is recognized that the diagram corresponding to a simple electron loop in Eq. (17) has no small  $(A/\mu)$ . The three diagrams that follow correspond to renormalization of a bare vertex and, as shown in Ref. 9, they can also be disregarded if  $A/\mu \ll 1$ . One remark is in order there. Diagrams that are rigorously zero for an easy-axis FM on account of conservation of the angular-momentum projection at the vertex can become nonzero in our case. At low values of the anisotropy constants  $D$  they acquire an additional smallness  $\sim (D/A\sigma)^2$ . For an anisotropy energy comparable with the molec-

ular-field energy, there remains in the diagrams only the small parameter  $(A/\mu)$ . Analogously, small quantities of order  $(A/\mu)$  are contained also in other diagrams for  $M$ . In the region where  $T \ll T_c$  and  $A/\mu \ll 1$  we can therefore put  $M = 0$  for the spin-wave excitation spectrum. The resultant approximation corresponds to the random-phase approximation generalized to include the case of SA of arbitrary symmetry, while the elementary-excitation spectrum of an anisotropic  $s$ - $f$  ferromagnet is described, as seen from (21), by the poles of the Green's function  $D_{\alpha\beta}^{(0)}(k)$ .

The solution of Eq. (17) for the function  $D_{\alpha\beta}^{(0)}$  is given in the Appendix. We write down here the dispersion equation (A8) for the collinear case frequently realized in practice, where the external magnetic field and the magnetic moment of the localized subsystem have the same direction. Using Eqs. (14) and (A5) we find from (A8) that the spin-wave spectrum satisfies the equation

$$\Delta(\mathbf{k}, \omega_n) = \begin{vmatrix} \Delta_{\parallel}(\mathbf{k}, \omega_n), & \Delta_{12}(\mathbf{k}, \omega_n) \\ \Delta_{21}(\mathbf{k}, \omega_n), & \Delta_{\perp}(\mathbf{k}, \omega_n) \end{vmatrix} = 0, \quad (22)$$

where

$$\Delta_{\parallel}(\mathbf{k}, \omega_n) = \begin{vmatrix} 1 - L^{zz}(\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & L^{zz}(\omega_n) \Pi_{\downarrow\downarrow}(\mathbf{k}, \omega_n) \\ L^{zz}(\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & 1 - L^{zz}(\omega_n) \Pi_{\downarrow\downarrow}(\mathbf{k}, \omega_n) \end{vmatrix},$$

$$\Delta_{\perp}(\mathbf{k}, \omega_n)$$

$$= \begin{vmatrix} 1 - L^{+-}(-\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & -L^{--}(\omega_n) \Pi_{\uparrow\downarrow}(\mathbf{k}, \omega_n) \\ -L^{++}(\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & 1 - L^{+-}(\omega_n) \Pi_{\uparrow\downarrow}(\mathbf{k}, \omega_n) \end{vmatrix}$$

$$\Delta_{12}(\mathbf{k}, \omega_n) = \begin{vmatrix} -L^{z+}(\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & -L^{z-}(\omega_n) \Pi_{\uparrow\downarrow}(\mathbf{k}, \omega_n) \\ L^{z+}(\omega_n) \Pi_{\uparrow\downarrow}(\mathbf{k}, \omega_n), & L^{z-}(\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n) \end{vmatrix}$$

$$\Delta_{21}(\mathbf{k}, \omega_n) = \begin{vmatrix} -L^{z-}(-\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & L^{z-}(-\omega_n) \Pi_{\downarrow\downarrow}(\mathbf{k}, \omega_n) \\ -L^{z+}(-\omega_n) \Pi_{\uparrow\uparrow}(\mathbf{k}, \omega_n), & L^{z+}(-\omega_n) \Pi_{\downarrow\downarrow}(\mathbf{k}, \omega_n) \end{vmatrix}.$$

The functions  $L(\omega_n)$  in these expressions are defined by the relations

$$L^{zz}(\omega_n) = \sum_{\alpha} |\gamma_{\parallel}(\alpha)|^2 D_{\alpha}(\omega_n) b(\alpha),$$

$$L^{z+}(\omega_n) = \sum_{\alpha} \gamma_{\parallel}(\alpha) \gamma_{\perp}(-\alpha) D_{\alpha}(\omega_n) b(\alpha),$$

$$L^{z-}(\omega_n) = \sum_{\alpha} \gamma_{\parallel}(\alpha) \gamma_{\perp}^*(\alpha) D_{\alpha}(\omega_n) b(\alpha),$$

$$L^{--}(\omega_n) = \sum_{\alpha} \gamma_{\perp}^*(\alpha) \gamma_{\perp}(-\alpha) D_{\alpha}(\omega_n) b(\alpha),$$

$$L^{++}(\omega_n) = \sum_{\alpha} \gamma_{\perp}(\alpha) \gamma_{\perp}(-\alpha) D_{\alpha}(\omega_n) b(\alpha),$$

$$L^{+-}(\omega_n) = \sum_{\alpha} |\gamma_{\perp}(\alpha)|^2 D_{\alpha}(\omega_n) b(\alpha). \quad (23)$$

We shall analyze the solutions of (22) for uniaxial and cubic ferromagnet metals.

#### 4. UNIAXIAL FERRIMAGNETIC METALS

In the analysis of the magnetic-excitation spectrum of this class of ferromagnets we confine ourselves to the collinear case, when the external magnetic field and the magneti-

zation have the same direction. We orient the field  $\mathbf{H}$  either along or across any anisotropy axis. We begin with the second case.

### a) $\mathbf{H}$ perpendicular to the anisotropy axis

We orient  $z$  along  $\mathbf{H}$  and  $\mathbf{M}$  and direct  $x$  along the anisotropy axis. We write the single-ion-anisotropy Hamiltonian in the form

$$\mathcal{H}_a(f) = 2D(S_z^x)^2.$$

This collinear case takes place if  $D > 0$  (easy-plane anisotropy) for any external magnetic field, or else if  $D < 0$  (easy-axis anisotropy) but in a sufficiently strong external magnetic field (see below).

From the solution of the one-ion problem (9) and from the definition of the parameters (10) it follows that  $\gamma_1(\alpha)\gamma_{||}(\alpha) = 0$  in this geometry. Therefore  $L^{z\pm}(\omega) = 0$  and the dispersion equation (22) breaks up into two separate equations for the spectra of the longitudinal and transverse oscillations of the magnetization.

We calculate first the longitudinal-oscillation spectrum of a uniaxial  $s$ - $f$  ferromagnet in a "planar" geometry. It follows from (22) that this spectrum is obtained by solving the equation

$$1 - L^{zz}(\omega) [\Pi_{\uparrow\uparrow}(\mathbf{k}, \omega) + \Pi_{\downarrow\downarrow}(\mathbf{k}, \omega)] = 0. \quad (24)$$

In the long-wave region, when  $|\mathbf{k}| \lesssim 2k_F\omega/\mu$  ( $\mu$  is the electron-gas Fermi energy and  $k_F$  is the Fermi quasimomentum), assuming a quadratic dispersion law for the electrons, calculation of the polarization operator  $\Pi_{\sigma\sigma}(\mathbf{k}, \omega)$  accurate to terms quadratic in the quasimomentum leads to the expression

$$\Pi_{\sigma\sigma}(\mathbf{k}, \omega) = \frac{A^2 n_\sigma}{m\omega^2} \mathbf{k}^2, \quad n_\sigma = \frac{1}{N} \sum_{\mathbf{p}} \left[ \exp\left(\frac{\varepsilon_{\mathbf{p}\sigma} - \mu}{T}\right) + 1 \right]^{-1}. \quad (25)$$

We take two circumstances into account in the calculation of the function  $L^{zz}(\omega)$ . First, we consider hereafter the specific spectral properties of  $s$ - $f$  magnets only at low temperatures,  $T \ll T_c$ . This allows us to neglect the contribution from transitions between upper one-ion states, whose populations are exponentially small. Second, we confine ourselves to ferromagnets that satisfy the relation

$$D \ll \bar{H} = 2A\sigma + g\mu_B H.$$

We use the letter  $\sigma$  to designate the spin variable as well as the magnetization of the itinerant electrons:  $\sigma = (n_\uparrow - n_\downarrow)/2$ .

Accurate to terms quadratic in  $(D/H)$  we have

$$L^{zz}(\omega) = \frac{2J(2J-1)\varepsilon_{31}}{\omega^2 - \varepsilon_{31}^2} \left(\frac{D}{\bar{H}}\right)^2, \quad (26)$$

where  $\varepsilon_{31} = 2H + 4(J-1)D$ .

Using the expressions obtained for the polarization operator  $\Pi_{\sigma\sigma}(\mathbf{k}, \omega)$  and for  $L^{zz}(\omega)$ , we obtain from (24) the dispersion law for the longitudinal branch:

$$\omega_{||}(\mathbf{k}) = \varepsilon_{31} + \frac{J(2J-1)}{6\pi^2} (3\pi^2 n)^{1/3} \left(\frac{A}{\bar{H}}\right)^2 \left(\frac{D}{\bar{H}}\right)^2 \mu \Omega_0^{2/3} \mathbf{k}^2, \quad (27)$$

where  $\Omega_0$  is the unit-cell volume and  $n = n_\uparrow + n_\downarrow$  is the normalized itinerant-electron density per magnetoactive ion.

The transverse-oscillations spectrum of an easy-plane  $s$ - $f$  ferromagnet (or an easy-axis one but in a strong enough transverse magnetic field) is determined, as follows from (22), by the solutions of the equation

$$\begin{aligned} [1 - L^{+-}(\omega)\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega)] [1 - L^{+-}(-\omega)\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega)] \\ - L^{++}(\omega)L^{--}(\omega)\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega)\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega) = 0. \end{aligned} \quad (28)$$

This equation describes, generally speaking, many spectrum branches of the elementary excitations due to collectivization of the transitions between one-ion states. In the low-temperature region, just as for the longitudinal-oscillation spectrum, we can neglect transitions between excited one-ion states. We obtain then from the solution of the one-ion problem and from Eqs. (10) and (23), accurate to terms  $\sim (D/H)^2$  inclusive,

$$L^{+-}(\omega) = \frac{2J}{\omega - \varepsilon_{21}} + J(2J-1) \frac{2(J-1)\varepsilon_{21} - \omega}{\omega^2 - \varepsilon_{21}^2} \left(\frac{D}{\bar{H}}\right)^2, \quad (29)$$

$$L^{++}(\omega)L^{--}(\omega) = \left(\frac{2J\varepsilon_{21}}{\omega^2 - \varepsilon_{21}^2}\right)^2 \frac{(2J-1)^2 D^2}{\bar{H}^2}, \quad (30)$$

where  $\varepsilon_{nm} = E_n - E_m$  is the difference between the energies of the one-ion states. By retaining the terms of second order in  $D/H$  we can obtain in explicit form the quantum corrections to the spectrum.

In the long-wave region, the analytic expression for the polarization spectrum can take the form of a series in powers of the quasimomentum. Using a quadratic dispersion law for the conduction electrons, we get

$$\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega) = \frac{2A^2\sigma}{\omega - \Delta} + \frac{(3\pi^2 n)^{1/3}}{3\pi^2} \mu A^2 \frac{\omega - \Delta x^2}{(\omega - \Delta)^3} \Omega_0^{2/3} \mathbf{k}^2, \quad (31)$$

where  $x = \Delta/4\pi$  and  $\Delta = 2\mu_B H + 2A(J^z)$ . This expansion is valid under the condition

$$|\omega - \Delta|/2\mu \gg |\mathbf{k}|/k_F. \quad (32)$$

An expansion for  $\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega)$  is obtained from (31) by recognizing that

$$\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega) = \Pi_{\uparrow\downarrow}(\mathbf{k}, -\omega).$$

Substituting (29)–(31) in the dispersion equation (28) and solving the latter, we find that the low-frequency spectrum branch of an easy-plane  $s$ - $f$  ferromagnet in collinear geometry is described by the equation

$$\omega^2(\mathbf{k}) = [\tilde{g}\mu_B H + c\Omega_0^{1/3}\mathbf{k}^2] [\tilde{g}\mu_B H + 2\bar{D} + c\Omega_0^{1/3}\mathbf{k}^2] + \delta^2, \quad (33)$$

where the  $g$  factor renormalized by the conduction electrons is

$$\tilde{g} = (gJ + 2\sigma)/(J + \sigma). \quad (34)$$

It is interesting to note that in the particular case  $g = 2$  no renormalization of the  $g$  factor takes place. The absence, at  $g = 2$ , of renormalization for isotropic  $s$ - $f$  ferromagnets was noted earlier (see, e.g., Ref. 5). The "phenomenological" anisotropy constant  $\bar{D}$  contains both the usual quantum renormalization and the renormalization due to free electrons:

$$\bar{D} = J(2J-1)D/(J+\sigma). \quad (35)$$

The spin-wave rigidity is defined by the expression

$$c = \frac{(3\pi^2 n)^{1/2}}{24\pi^2} \left( \frac{J}{J+\sigma} \right) \left( \frac{A^2 J}{\mu} \right) \left[ 1 - \frac{2\omega_0^2}{AJ} \frac{(\mu/AJ)^2}{\tilde{g}\mu_B H + D} \right]. \quad (36)$$

The gap in the spectrum of the considered branch is equal to

$$\omega_0 = \omega(\mathbf{k}=0) = (\omega_{cl}^2(0) + \delta^2)^{1/2}, \quad (37)$$

where  $\omega_{cl}(0)$  has the usual phenomenological form for a uniaxial ferromagnet in transverse geometry:

$$\omega_{cl}(0) = (\tilde{g}\mu_B H (\tilde{g}\mu_B H + 2D))^{1/2}, \quad (38)$$

and the additional term

$$\delta^2 = \frac{2J(2J-1)}{J+\sigma} \left( \frac{\tilde{g}\mu_B H}{H} \right) D^2 \quad (39)$$

is due to quantum corrections. Such corrections for the elementary-excitation spectrum were predicted earlier in the theory of anisotropic and multisublattice Heisenberg magnets.<sup>21,22</sup> Allowance for zero-point quantum oscillations leads to a higher activation energy of the spin-wave excitations.

It follows from (37) that at  $D < 0$  our analysis is valid for fields

$$H > \frac{2J(2J-1)}{\tilde{g}\mu_B(J+\sigma)} \left[ |D| - \frac{D^2}{H} \right].$$

The second term in the square brackets determines the quantum renormalization of the critical field with respect to an orientational transition.

It follows from (28) that the expression for the gap in the spectrum of the upper branch is

$$\Omega = 2|A|(J+\sigma) + \frac{A}{|A|} \left[ \frac{g\sigma + 2J}{J+\sigma} \mu_B H + \frac{\sigma(2J-1)}{J+\sigma} D \right]. \quad (40)$$

Let us discuss the peculiarities of the dispersion law for spin-wave excitations of  $s$ - $f$  ferromagnets in the considered geometry. It follows from (36) that the second term in the square brackets is a product of a small quantity by the square of a large parameter, since the approximation employed implies that  $\mu \gg |A|J$ . For  $H = 0$  we have from the Goldstone theorem  $\omega_0 = 0$  and the second term vanishes. This term can also be disregarded for finite magnetic fields, subject to the inequality

$$\frac{2\omega_0^2}{|A|J} \frac{(\mu/AJ)^2}{\tilde{g}\mu_B H + D} \ll 1. \quad (41)$$

The dependence of the frequency of the long-wave excitations on the field  $H$ , on the anisotropy  $D$ , and on the quasimomentum  $\mathbf{k}$  will coincide then with the corresponding results of a theory based on the use of the Heisenberg model to describe exchange interaction between localized moments, followed by allowance for one-ion anisotropy and for Zeeman interaction. The exchange constants of the Heisenberg Hamiltonian are calculated from the usual RKKY theory. We designate hereafter, for brevity, this use of the RKKY theory with subsequent allowance for single-site interactions (SI) by RKKY + SI.

It can be stated in this connection that the inequality (41) is the condition under which the RKKY + SI model

can be used to describe the properties of long-wave excitations of metallic uniaxial ferromagnets in a transverse geometry.

If  $D > 0$ , condition (41) can be written in simpler form:

$$\frac{2\tilde{g}\mu_B H}{|A|J} \left( \frac{\mu}{AJ} \right)^2 \ll 1. \quad (42)$$

We have neglected here the fact that the terms  $\sim D$  in the expression for  $\omega_{cl}(0)$  and in Eq. (41) differ by the factor two. Therefore in the magnetic field region

$$H \ll H_0, \quad H_0 = \left( \frac{|A|J}{2\tilde{g}\mu_B} \right) \left( \frac{AJ}{\mu} \right)^2,$$

the spin-wave rigidity is independent of  $H$ . The character of the dispersion law changes greatly at  $H \sim H_0$ . This is most pronounced for positive  $A$ . In this case the frequency varies anomalously at  $H > H_0$ , and an increase of the quasimomentum decreases the excitation energy. The dispersion law has therefore a minimum at a certain quasimomentum value. Since an increase of the magnetic field leads to an increase of  $\omega_0$  it is obvious beforehand that a change of the dispersion for small  $\mathbf{k}$  does not mean instability of the ferromagnetic states, since  $0 < \omega_{\min} < \omega_0$ .

The onset and behavior of this anomaly is, naturally not confined to the considered easy-plane ferromagnet. The point is that the cause of the change of the spectral properties is the peculiar frequency dependence of the polarization operator. This operator is known to play the role of a Fourier transform of the exchange integral (this is easily verified in our case by comparing the dispersion equation (28) with the corresponding equation of the RKKY + SI model.<sup>15</sup> The polarization operator  $\Pi_{\perp}(\mathbf{k}, \omega)$  is therefore indicative, on the one hand, of the interaction between the localized moments, and defines, on the other, the dispersion properties of the spin-wave excitation spectrum. It is important here that the character of the dependence of  $\Pi_{\perp}(\mathbf{k}, \omega)$  on  $\mathbf{k}$  in the long-wave region is determined, as seen from (31), by that energy interval in which the excitations of interest to us are realized. Since the energy of excitations with  $\mathbf{k} = 0$  is determined by the external magnetic field, by the intensity of the single-ion anisotropy, and so on, variation of these fields entails also variation of the energy region in which long-wave excitations exist. It is owing to this peculiarity of the polarization operator that variation of the energy of an excitation with  $\mathbf{k} = 0$  influences also the character of the dispersion.

Naturally, since this mechanism is general, it is effective also in an easy-axis  $s$ - $f$  ferromagnet and others (see below). The quantitative aspect of the anomaly question is whether such excitation energies can be reached when  $\omega_0 \sim \Delta x^2$  and depends on the initial parameters of the system.

Let us demonstrate the described peculiarity of the transformation of a long-wave spectrum when the magnetic field is varied, using an isotropic  $s$ - $f$  ferromagnet as the example. The excitation spectrum can then be obtained from (33) by putting  $D = 0$ . We have

$$\omega(\mathbf{k}) = \tilde{g}\mu_B H + \frac{(3\pi^2 n)^{1/2}}{24\pi^2} \left( \frac{J}{J+\sigma} \right) \left( \frac{A^2 J}{\mu} \right) \times \left[ 1 - \frac{2\tilde{g}\mu_B H}{AJ} \left( \frac{\mu}{AJ} \right)^2 \right] \Omega_0^{1/2} \mathbf{k}^2. \quad (43)$$

This equation agrees with a known result<sup>5</sup> if  $g = 2$  and

$H = 0$ . To find the excitation energies outside the long-wave region we use a numerical solution of the dispersion equation for an isotropic  $s$ - $f$  ferromagnet<sup>5</sup>:

$$\omega = \varepsilon_{21} + 2J\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega), \quad \varepsilon_{21} = g\mu_B H + 2A\sigma.$$

We choose in the numerical calculations the same values for the parameters of the rare-earth metals Gd, Tb, ..., Tm as in Refs. 5 and 6. We estimate here only the external magnetic field strength  $H_0$  starting with which the dispersion dependence becomes a function of  $H$ .

In pure rare-earth single-crystal Gd, Tb, ..., Tm, the RE ions give up three electrons each to the collectivized states. In the simplest estimates<sup>5,6</sup> one assumes for these metals  $n = 3$ , an atomic volume  $\Omega_0 = 18 \text{ cm}^3 \cdot \text{mol}^{-1}$ , and  $m = 3m_0$ , so that  $\mu = 2.63 \text{ eV}$ . This choice of the model parameters ensures satisfactory agreement of the experimental and theoretical values of the paramagnetic Curie temperatures  $\Theta$  for the rare-earth metals (see Fig. 20.35 of Ref. 5) if the de Gennes factor<sup>6</sup> is taken into account, i.e., if we substitute  $A \rightarrow A = A_{sf}(g - 1)$  and choose  $A_{sf} = 0.19 \text{ eV}$ .<sup>5</sup> Using these data we get  $H_0 = 1.83 \cdot 10^6 \text{ Oe}$  for single-crystal Gd ( $J = 7/2$  and  $g = 2$  for  $\text{Gd}^{3+}$ ). Such fields are difficult to produce, and it is therefore difficult to change the spin-wave rigidity of metallic Gd by using a magnetic field. The situation for single-crystal Tm, however, is already different. Recognizing that  $J = 6$  and  $g = 7/6$  for Tm, we obtain, at the same values of the remaining parameters,  $H_0 = 73.3 \text{ kOe}$ . In strong magnetic fields, Tm is a collinear ferromagnet,<sup>6</sup> and the anomaly induced in the spectrum by the field  $H$  can be investigated experimentally.

Figure 1 shows the dependence of the frequency on the wave vector for ferromagnet Tm without allowance for the anisotropy at  $H = 0$  (curve 1) and in a field  $H = 100 \text{ kOe}$  (curve 2). The abscissas are the values of the wave vector in units of  $(2m\Delta)^{1/2}$ . The "boundary" value of the quasimomentum  $k = \pi/a$  corresponds here to  $q = q_b = 1.85$ . The ordinates are the frequencies reckoned from  $\omega_0 = \tilde{g}\mu_B H$ , measured in units of the width  $W$  of the spin-wave band. The quantity  $W$  is defined as the excitation frequency in a field  $H = 0$  at the "boundary" of the band, i.e.,  $W$  is obtained by solving the dispersion equation with  $q = q_b$ , namely  $W = (k = \pi/a)$ . In our case  $W = 5.07 \text{ K}$ . It is seen from the figure that in a zero external field the dispersion curve has only an insignificant anomaly in the dispersion curve at wave vector values  $q \approx (\Delta/\mu)^{1/2}/2$ . In dimensional units we find that the anomaly occurs, as usual, at quasimomentum values

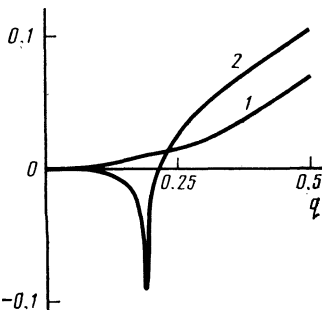


FIG. 1. Spectrum of magnetic excitations of the lower branch of an isotropic  $s$ - $f$  ferromagnet at two values of the external magnetic field: 1— $H = 0$ , 2— $H = 100 \text{ kOe}$ .

$k \approx \Delta(m/2\mu)^{1/2} \approx p_{\uparrow} - p_{\downarrow}$ , where  $p_0$  is the limiting Fermi momentum for a subband with spin projection  $\sigma$ .

In a field  $H = 100 \text{ kOe}$  the behavior of the excitation frequency changes radically with increase of the quasimomentum. For small quasimomenta the spin-wave rigidity is negative, with a minimum in the region  $k = p_{\uparrow} - p_{\downarrow}$ . This anomaly of the behavior of the frequency as a function of the quasimomentum is induced by the magnetic field and cannot be described in the framework of the RKKY + OI model.

Qualitatively similar results are obtained also with other parameters of the model. The described anomaly will therefore be induced by a magnetic field also in other  $s$ - $f$  ferromagnets, such as intermetallides. The experiments should be performed with ferromagnetic compounds that are magnetically ordered at low temperatures. The field  $H_0$  is then easily attainable in experiment.

If  $A < 0$ , an increase of  $H$  does not reverse the sign of the spin-wave rigidity. However, in this case too the spin-wave rigidity becomes dependent on the field and on the anisotropy constants, starting with a field  $H \sim H_0$ .

### b) $H$ parallel to the anisotropy axis

For this geometry we align the  $z$  axis with the crystal axis. We express the one-ion anisotropy Hamiltonian as an arbitrary function of the operator  $S_z^2$

$$\mathcal{H}_a(f) = -\varphi(S_z^2). \quad (44)$$

It is assumed that the conditions that ensure collinear geometry of the problem are met (see below). The dispersion equation is

$$[1 - L^{+-}(\omega)\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega)][1 - L^{+-}(-\omega)\Pi_{\uparrow\downarrow}(\mathbf{k}, \omega)] = 0 \quad (45)$$

and has been obtained earlier in Ref. 9. In the low-temperature region, the spectrum of the lower branch of the long-wave oscillations of the magnetization is given by

$$\omega(\mathbf{k}) = \omega_0 + c\Omega_0^{3/2}k^2, \quad (46)$$

where the spin-wave rigidity  $c$  is of the form

$$c = \frac{(3\pi^2 n)^{1/2}}{24\pi^2} \left( \frac{J}{J+\sigma} \right) \left( \frac{A^2 J}{\mu} \right) \left[ 1 - \left( \frac{2\omega_0}{AJ} \right) \left( \frac{\mu}{AJ} \right)^2 \right], \quad (47)$$

and the gap in the spectrum is  $\omega_0 = \tilde{g}\mu_B H + J\varepsilon_a/(J + \sigma)$ , where  $\varepsilon_a = \varphi(J) - \varphi(J - 1)$ . The frequency of the upper branch is given at  $\mathbf{k} = 0$  by

$$\Omega = 2|A|(J + \sigma) + \frac{A}{|A|} \left[ \frac{g\sigma + 2J}{J + \sigma} \mu_B H + \frac{\sigma}{J + \sigma} \varepsilon_a \right]. \quad (48)$$

Our reasoning is correct if  $\omega_0 > 0$ . The foregoing analysis is therefore valid for easy-axis ferromagnets also at  $H = 0$ , whereas if  $\varepsilon_a < 0$  the magnetic field should exceed a certain critical value. It is seen from (46) that the conduction electrons renormalize the energy of the single-ion anisotropy.

Just as in the preceding cases, strengthening the magnetic field can influence the spin-wave rigidity of the system. The difference in the geometry considered here is that the one-ion anisotropy energy enters additively in the expression for  $\omega_0$ . Thus, for  $\varphi(M) = DM^2$  we have

$$\omega_0 = \tilde{g}\mu_B H + \left( \frac{J}{J + \sigma} \right) (2J - 1)D.$$

It follows hence that in easy-axis ferromagnets ( $D > 0$ ) the field  $H_0$ , starting with which a strong anomaly appears in the dispersion law for the lower branch, becomes weaker. This anomaly can occur therefore in a strongly anisotropic ferromagnet also at  $H = 0$ . Thus, the theory of strongly anisotropic easy-axis ferromagnets, when

$$D \gg \left(\frac{2J-1}{2}\right) |A| J \left(\frac{AJ}{\mu}\right)^2,$$

cannot be consistently considered in the framework of the RKKY + SI model, for in the latter approach an increase of the anisotropy leads only to a shift of the entire spin-wave spectrum (towards longer wavelengths), without changing the character of the dependence of the frequency on the wave vector. Actually, however, at large  $D$  the spin-wave rigidity becomes dependent on the anisotropy. This means in fact that the exchange interaction that sets in between the RE ions in strongly anisotropic metallic ferromagnets is determined not only by the properties of the electron subsystem, but also by the character of the nonequidistant one-ion energy levels.

## 5. METALLIC FERROMAGNETS WITH CUBIC-SYMMETRY ONE-ION ANISOTROPY

Consider a metallic ferromagnet in which the magnetoactive ions are in a cubic-symmetry crystal field. Examples of such systems are monochalcogenides of uranium,<sup>3</sup> and some intermetallic compounds of the  $\text{ReAl}_2$  family.<sup>1,2</sup> If the quantization axis is aligned with one of the fourfold axes, the Hamiltonian of the single-ion anisotropy takes the form

$$\mathcal{H}_a(f) = \frac{Wx}{F_4} [O_4^0 + 5O_4^4] + \frac{W(1-|x|)}{F_6} [O_6^0 - 21O_6^4]. \quad (49)$$

This parametrization of the crystal-field energy operator was introduced in Ref. 23 and is constantly used to describe the physical properties of RE compounds with cubic anisotropy.<sup>1,2,24</sup> The energy parameter  $W$  specifies the splitting of the multiplet level by the crystal field. The parameter  $x$  can take on values in the range from  $-1$  to  $+1$  and determines the relative contribution made to the anisotropy energy by invariants of fourth and sixth order. The actual values of  $W$  and  $x$  specify the direction of the easy-magnetization axis (EMA) and depends therefore both on the RE ion (in the structurally isomorphous compounds ( $\text{DyAl}_2$ ,  $\text{HoAl}_2$ , and  $\text{TbAl}_2$ , for example, the EMA at low temperatures coincides with the directions  $[100]$ ,  $[110]$ , and  $[111]$ , respectively), and on the type of compound (the moments of the  $\text{Ho}^{3+}$  ion in ferromagnetically ordered  $\text{HoP}$  are oriented along the  $[100]$  direction). The ranges of the parameters  $W$  and  $x$  in which a phase with a specified EMA axis exists, at least in a metastable state, is determined from the condition that the elementary-excitation spectrum be positive-definite. The coefficients  $F_4$  and  $F_6$  depend on the values of the angular momentum  $J$ .<sup>2</sup>

We begin the analysis of cubic ferromagnet metals with the case when the easy magnetization axis is  $C_4$ . Confining ourselves to the collinear case, we assume that the external magnetic field is also oriented along the fourfold axis.

### a) EMA $\parallel [100] \parallel H$

In this geometry, the one-ion Hamiltonian is obtained from (49) by adding a term  $-\bar{H}J^z$  with an effective field  $\bar{H} = g\mu_B H + 2A$ . From an analysis of the properties of the wave functions of one-ion states<sup>25</sup> it follows that  $\gamma_{\perp}(\alpha)\gamma_{\perp}(-\alpha) = 0$ ,  $\gamma_{\parallel}(\alpha)\gamma_{\perp}(-\alpha) = 0$ . We find therefore that the spectrum of the transverse oscillations is described by an equation that coincides formally with (45), while the spectrum of the longitudinal oscillations is described by Eq. (24). These dispersion equations make it possible to investigate the spectral properties of metallic ferromagnets at sufficiently high one-ion anisotropy energies. The entire manifestation of the quantum effects can then be traced. Unfortunately, owing to the large values of  $J$ , it is impossible to solve analytically the one-ion problem for real ferromagnets. Numerical methods are therefore used. There is nonetheless a parameter range in which the SA energy is lower than the splitting due to the field  $H$ , but at the same time high enough for the quantum effects to become noticeable. In this case the one-ion problem can be solved by standard perturbation theory. Inclusion of terms of second order in  $(W/\bar{H})$  yields the first-order quantum corrections. The analysis is carried out for arbitrary  $J$ .

Thus, an investigation of the transverse-oscillation spectrum shows that at  $I \ll T_c$  we have

$$L^{+-}(\omega) = \frac{\gamma_{\perp}^2(12)}{\omega - \epsilon_{21}} + \frac{\gamma_{\perp}^2(16)}{\omega - \epsilon_{61}} - \frac{\gamma_{\perp}^2(41)}{\omega + \epsilon_{41}}. \quad (50)$$

It follows from this equation and from (45) that in the given geometry a cubic anisotropy leads to collectivization of the excitations due to transitions between the states  $|\Psi_6\rangle$  and  $|\Psi_1\rangle$ , and also between  $|\Psi_4\rangle$  and  $|\Psi_1\rangle$ . The characteristic energies of such excitations are  $\omega \approx \omega_{61}$  and  $\omega \approx \omega_{41}$ , respectively, and the bandwidth is  $\sim W^2/\bar{H}$  and is small since the anisotropy is small.

Solving Eq. (45), we find that the gap in the optical-branch spectrum is given by Eq. (48) in which we substitute

$$\epsilon_a \rightarrow \epsilon_{100} = -\left(\frac{2WK_J}{F_4}\right) [10x + 21C_J(1-|x|)].$$

The coefficient  $K_J = (2J-1)(J-1)(2J-3)$  takes into account the "kinematic" renormalization of the anisotropic constant and show in explicit form that there is no cubic anisotropy for  $J = 1/2$ ,  $1$ , and  $3/2$ . The quantity  $C_J = (J-2)(2J-5)(F_4/F_6)$  vanishes at  $J = 2$  and  $5/2$ , since there is no sixth-order invariant for these values of  $J$ .

It follows from the expression for the optical-branch gap that the anisotropy contribution is subject to substantial renormalization. The reason is that the optical branch of the excitation is due to Stoner transitions of the conduction electrons, which by themselves are unaffected by the anisotropy. On the other hand, the term connected with the anisotropy is governed by the extent to which the itinerant and localized electrons are coupled. Since the SA plays a substantial role for the latter, the anisotropy effects are transferred via the  $s$ - $f$  exchange coupling to the collectivized subsystem and are manifested, in particular in the excitation energies of the itinerant electrons.

The spectrum of the long-wave excitations of the lower branch of the transverse oscillations is described by Eq. (46), except that  $\omega_0$  must be understood to mean



$$\omega_0 = \tilde{g}\mu_B H + \left(\frac{J}{J+\sigma}\right) \varepsilon_{100} + \Delta\omega_{qu}. \quad (51)$$

Here  $\Delta\omega_{qu}$  is due to quantum effects:

$$\Delta\omega_{qu} = \left(\frac{J}{J+\sigma}\right) \left(\frac{K_J}{H}\right) \left(\frac{10W}{F_4}\right)^2 \{Jf^2(x) - 3(J-2)[2\varphi^2(x) + f(x)\varphi(x)]\},$$

where

$$f(x) = x - 21C_J(1-|x|), \quad \varphi(x) = f(x) + \frac{231}{5} \left(\frac{C_J}{J-2}\right) (1-|x|)$$

The constraint on  $W$  for each  $J$  follows from the condition for the validity of our analysis when  $|\Delta\omega_{qu}| \ll \omega_0$ . Just as before, an anomaly appears in the spectrum of the considered branch if

$$\frac{2\omega_0}{|A|J} \left(\frac{\mu}{AJ}\right)^2 \gg 1. \quad (52)$$

Let us find the long-wave spectrum of the longitudinal oscillations. At the stipulated accuracy we have

$$L^{zz}(\omega) = \left(\frac{48JK_J}{H}\right) \left(\frac{10W}{F_4}\right)^2 \frac{f^2(x)}{\omega^2 - \varepsilon_{51}^2}. \quad (53)$$

Using expression (25) for the polarization operator  $\Pi_{\sigma\sigma}(\mathbf{k}, \omega)$  for small quasimomentum values, we get from (24) the desired dispersion relation

$$\omega_{||}(\mathbf{k}) = \varepsilon_{51} + JK_J \frac{(3\pi^2 n)^{1/2}}{\pi^2} \left[ \frac{5WAf(x)}{F_4 H^2} \right]^2 \mu \Omega_0^{3/2} \mathbf{k}^2. \quad (54)$$

Let us analyze the excitation spectrum of a cubic metallic ferromagnet with a threefold axis as the easy magnetization axis.

#### b) EMA $||[111]||H$

In this geometry, the  $z$  axis is aligned with the  $C_3$  axis. The form of the cubic-anisotropy operator for this choice of the quantization axis is well known.<sup>24</sup> Just as in the preceding case, the excitation spectrum is determined by Eqs. (24) and (45).

The dispersion relation for the lower branch of transverse oscillations is described by Eq. (46) in which the gap is given by

$$\begin{aligned} \omega_0 &= \tilde{g}\mu_B H + \left(\frac{J}{J+\sigma}\right) \left(\frac{8WK_J}{3F_4}\right) [5x - 28C_J(1-|x|)] + \Delta\omega_{qu}, \\ \Delta\omega_{qu} &= -\left(\frac{J}{J+\sigma}\right) \left(\frac{2K_J}{H}\right) \left(\frac{40W}{3F_4}\right)^2 \left\{ \frac{(J-3)(18J-35)}{4} x^2 \right. \\ &\quad + 7 \left(6J^2 - \frac{133}{3}J + 68\right) \\ &\quad \times C_J x(1-|x|) - 49 \left[ \left(\frac{11}{25}\right)^2 \frac{2(10-3J)F_4}{F_6} \right. \\ &\quad \left. - \left(2J^2 - \frac{59}{3}J + \frac{175}{4}\right) C_J \right] \\ &\quad \left. \times C_J (1-|x|)^2 \right\}. \end{aligned}$$

We obtain an equation for the optical-branch gap from (48) by making the substitution

$$\varepsilon_a \rightarrow \varepsilon_{111} = \left(\frac{8WK_J}{3F_4}\right) [5x - 28C_J(1-|x|)].$$

It follows from the analysis of the one-ion problem that

$$L^{zz}(\omega) = \frac{2\gamma_{||}^2(14)\varepsilon_{41}}{\omega^2 - \varepsilon_{41}^2} + \frac{2\gamma_{||}^2(17)\varepsilon_{71}}{\omega^2 - \varepsilon_{71}^2}.$$

The spectrum of the longitudinal oscillations, in contrast to the preceding case, consists of two branches; the dispersion relation for the lower branch is

$$\begin{aligned} \omega_{||}^{(1)}(\mathbf{k}) &= \varepsilon_{41} + \frac{8(3\pi^2 n)^{1/2}}{\pi^2} J(2J-3)K_J \left(\frac{20WA}{9H^2 F_4}\right)^2 \\ &\quad \times \left[ x + \frac{14}{3} C_J(1-|x|) \right]^2 \mu \Omega_0^{3/2} \mathbf{k}^2. \end{aligned}$$

The dispersion equation for the upper-branch oscillations is described by the equation

$$\begin{aligned} \omega_{||}^{(2)}(\mathbf{k}) &= \varepsilon_{71} + \frac{5(3\pi^2 n)^{1/2}}{3\pi^2} J(J-2)(2J-5)K_J \\ &\quad \times \left[ \frac{8.77AW(1-|x|)}{9F_6 H^2} \right]^2 \mu \Omega_0^{3/2} \mathbf{k}^2. \end{aligned}$$

#### c) EMA $||[110]||H$

The form of the SA energy operator in the case when the quantization axis coincides with the  $[110]$  direction is given in Ref. 24. The difference between this geometry and the two previously considered is manifested, in particular by the fact that  $L^{++}(\omega)$  and  $L^{--}(\omega)$  do not vanish. The spectrum of the transverse oscillations is therefore described by the solutions of Eq. (28). Calculations by the described procedure yield the spin-wave spectrum of the lower branch of the transverse oscillations (disregarding the quantum corrections for the gap)

$$\begin{aligned} \omega^2 &= \left\{ \tilde{g}\mu_B H + \left(\frac{J}{J+\sigma}\right) \left(\frac{2WK_J}{F_4}\right) [10x + 21C_J(1-|x|)] \right. \\ &\quad \left. + c_{110} \Omega_0^{3/2} \mathbf{k}^2 \right\} \\ &\quad \times \left\{ \tilde{g}\mu_B H + \left(\frac{J}{J+\sigma}\right) \left(\frac{WK_J}{F_4}\right) \left[ -10x + \frac{189}{2} C_J(1-|x|) \right] \right. \\ &\quad \left. + c_{110} \Omega_0^{3/2} \mathbf{k}^2 \right\}, \quad (55) \end{aligned}$$

where

$$\begin{aligned} c_{110} &= \left(\frac{J}{J+\sigma}\right) \frac{(3\pi^2 n)^{1/2}}{24\pi^2} \left(\frac{A^2 J}{\mu}\right) \left[ 1 - \left(\frac{2\omega_0^2}{AJ}\right) \frac{(\mu/AJ)^2}{\tilde{g}\mu_B H + \varepsilon_{110}} \right], \\ \varepsilon_{110} &= \left(\frac{J}{J+\sigma}\right) \left(\frac{WK_J}{F_4}\right) \left\{ 5x + \frac{21 \cdot 13}{4} C_J(1-|x|) \right\}. \quad (56) \end{aligned}$$

In a zero external magnetic field, the region of existence of a ferromagnetic state with the magnetic moment directed along a twofold axis is defined by the conditions

$$W > 0, \quad -\frac{21C_J}{10+21C_J} < \tau < \frac{189C_J}{20+189C_J}.$$

The spectrum of the longitudinal oscillations consists of three branches:

$$\begin{aligned}\omega_{\parallel}^{(1)}(\mathbf{k}) &= \varepsilon_{s1} + \frac{3}{2}J(J-1)(2J-3)K_J(3\pi^2n)^{1/2} \left( \frac{5AW}{\pi F_s \bar{H}^2} \right)^2 \\ &\quad \times \left[ x - \frac{7}{4}C_J(1-|x|) \right]^2 \mu\Omega_0^{3/2}k^2, \\ \omega_{\parallel}^{(2)}(\mathbf{k}) &= \varepsilon_{s1} + JK_J(3\pi^2n)^{1/2} \left( \frac{15AW}{4\pi F_s \bar{H}^2} \right)^2 \\ &\quad \times \left[ x + \frac{35}{2}C_J(1-|x|) \right]^2 \mu\Omega_0^{3/2}k^2, \\ \omega_{\parallel}^{(3)}(\mathbf{k}) &= \varepsilon_{r1} + 15J(J-2)(2J-5)K_J(3\pi^2n)^{1/2} \\ &\quad \times \left[ \frac{77AW(1-|x|)}{4\pi F_s \bar{H}^2} \right]^2 \mu\Omega_0^{3/2}k^2.\end{aligned}\quad (57)$$

It follows from the results that in the long-wave region of the spectrum the spin-wave rigidity of the acoustic branch of cubic metallic ferromagnets depends on  $W$ ,  $x$ , and  $H$  if  $\omega_0 \gtrsim \Delta(\Delta/4\mu)^2$ .

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## APPENDIX

To find  $D_{\alpha\beta}^{(0)}$  we rewrite Eq. (17) in analytic form

$$\begin{aligned}D_{\alpha\beta}^{(0)}(k) &= \delta_{\alpha\beta} D_{\alpha}(\omega_n) \\ &\quad + D_{\alpha}(\omega_n) b(\alpha) \frac{T}{N} \sum_{\mathbf{p}, \alpha_1} \text{Sp} \{ \hat{G}(\mathbf{p}) \hat{\Gamma}_{\mathbf{k}}(-\alpha) \hat{G}(\mathbf{p}+\mathbf{k}) \\ &\quad \times \hat{\Gamma}_{\mathbf{k}}(\alpha_1) \} D_{\alpha\beta}^{(0)}(k).\end{aligned}\quad (A1)$$

Expanding the trace operation, we obtain in explicit form the summation over the spin indices. Summation over the intermediate frequencies leads to formation of the polarization operator:

$$\Pi_{\sigma\sigma'}(k) = \frac{A_{\mathbf{k}}}{N} \sum_{\mathbf{p}} \frac{n_{\sigma}(\mathbf{p}) - n_{\sigma'}(\mathbf{p}+\mathbf{k})}{i\omega_n + \varepsilon_{\sigma}(\mathbf{p}) - \varepsilon_{\sigma'}(\mathbf{p}+\mathbf{k})}, \quad (A2)$$

where  $n_{\sigma}(\mathbf{p}) = f_F \{ [\varepsilon_{\sigma}(\mathbf{p}) - \mu] / T \}$  is the Fermi-Dirac distribution function. To solve the system (A1), we introduce the following combination of the functions  $D_{\alpha\beta}^{(0)}(k)$ :

$$\Phi_{\beta}^{\sigma\sigma'}(k) = \sum_{\alpha} \Gamma^{\sigma\sigma'}(\alpha) D_{\alpha\beta}^{(0)}(k), \quad \Gamma^{\sigma\sigma'}(\alpha) = \Gamma_{\mathbf{k}}^{\sigma\sigma'}(\alpha) / A_{\mathbf{k}}.$$

It follows then from (A1) that  $D_{\alpha\beta}^{(0)}$  can be expressed in terms of  $\Phi_{\beta}^{\sigma\sigma'}$ :

$$\begin{aligned}D_{\alpha\beta}^{(0)}(k) &= \delta_{\alpha\beta} D_{\alpha}(\omega_n) + D_{\alpha}(\omega_n) b(\alpha) \\ &\quad \times \sum_{\sigma\sigma'} \Gamma^{\sigma\sigma'}(-\alpha) \Pi_{\sigma\sigma'}(k) \Phi_{\beta}^{\sigma\sigma'}(k).\end{aligned}\quad (A3)$$

Multiplying this equation by  $\Gamma^{\sigma\sigma'}(\alpha)$  and summing over  $\alpha$ , we obtain a closed system of four equations ( $\sigma, \sigma' = \pm \frac{1}{2}$  for each  $\beta$ ) for  $\Phi_{\beta}^{\sigma\sigma'}(k)$ :

$$\Phi_{\beta}^{\sigma\sigma'}(k) = \Gamma^{\sigma\sigma'}(\beta) D_{\beta} + \sum_{\sigma''} L_{\sigma\sigma''}^{\sigma\sigma'} \Pi_{\sigma\sigma''} \Phi_{\beta}^{\sigma''\sigma}, \quad (A4)$$

where

$$L_{\sigma\sigma''}^{\sigma\sigma'} = \sum_{\alpha} \Gamma^{\sigma\sigma'}(\alpha) \Gamma^{\sigma\sigma''}(-\alpha) D_{\alpha}(\omega_n) b(\alpha). \quad (A5)$$

Introducing a  $4 \times 4$  matrix  $W$  with elements

$$W_{\sigma\sigma', \sigma''\sigma''} = L_{\sigma\sigma''}^{\sigma\sigma'}(\omega_n) \Pi_{\sigma\sigma''}(\mathbf{k}, \omega_n), \quad (A6)$$

we get

$$\Phi_{\beta}^{\sigma\sigma'} = \sum_{\sigma_1\sigma_1'} \{ (I-W)^{-1} \}_{\sigma\sigma', \sigma_1\sigma_1'} \Gamma^{\sigma_1\sigma_1'}(\beta) D_{\beta}, \quad (A7)$$

where  $I$  is a unit matrix. Equations (A3) and (A7) determine the sought function  $D_{\alpha\beta}^{(0)}$ . The dispersion equation that describes the spectrum of the spin-wave excitations is

$$\Delta(k, i\omega_n \rightarrow \omega - i\delta) = \det \| I - W \| = 0. \quad (A8)$$

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