

Spin-Wave Dispersion Relation for Er Metal at 4.5°K*

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The spin-wave dispersion relation $\hbar\omega(\vec{q})$ for the c direction of erbium metal in its conical magnetic phase has been studied by coherent inelastic scattering of thermal neutrons. The improved resolution of the present experiment compared with that used in earlier work on Er has permitted the first observation of both the distinct spin-wave branches corresponding to $\hbar\omega(+\vec{q})$ and $\hbar\omega(-\vec{q})$ over a wide range of $|\vec{q}|$. As a consequence these results are the first to indicate that the anisotropic exchange interaction is a dominant interaction for a rare-earth metal and/or the model magnetic Hamiltonian generally employed for these metals is seriously inadequate.

The energies $\hbar\omega(\vec{q})$ of spin waves propagating along the c direction of erbium metal in its conical magnetic phase at 4.5°K have been measured by triple-axis neutron spectrometry. Although similar experimental work has been reported previously for Er,¹ Ho,² and an Er_{0.5}Ho_{0.5} alloy,³ we believe the present measurements are the first to resolve the two distinct spin-wave branches in a conical structure, which correspond to waves propagating parallel, $+\vec{q}$, and antiparallel, $-\vec{q}$, with respect to the ferromagnetic component of the magnetic moment, i.e., $\hbar\omega(+\vec{q}) \neq \hbar\omega(-\vec{q})$. Consequently a much more critical test of the theoretical model for the magnetic properties of Er can be made than would otherwise be possible.

The measurements were carried out on a crystal enriched (96.1%) in the low neutron capturing isotope ¹⁷⁰Er using a neutron spectrometer located at the Oak Ridge high-flux isotope reactor. Neutrons with energies in the range 11–15 meV generally were used. With Be crystals as monochromator and analyzer, and with moderate collimation before and after the sample [~ 20 min full width at half-maximum (FWHM)], the energy width (FWHM) of the instrumental resolution varied from 0.2 to 0.4 meV depending on the neutron energy and the energy transferred to the sample.

The neutron scattering cross section from spin-wave creation in a conical structure⁴ shows that for the neutron momentum transfer $\hbar\vec{Q}$ parallel to \vec{k}_0 , both of the Bragg-magnetic satellites at positions $2\pi\vec{\tau} \pm \vec{k}_0$ are origins of the spin-wave dispersion relation. Here $2\pi\vec{\tau}$ is a reciprocal lattice point, and \vec{k}_0 is a wave vector (in the c direction) which specifies the periodicity of the helical component of the structure. In a multi-domain sample the $\hbar\omega(+\vec{q})$ and $\hbar\omega(-\vec{q})$ branches are superimposed so that generally one expects to observe four peaks in the distribution of neutrons scattered under constant- \vec{Q} conditions.

However, the intensities expected for these branches can be quite different, and in addition there are several \vec{Q} 's, e.g., $(0, 0, 1)2\pi/c$, $(0, 0, 2) \times 2\pi/c$, and $(0, 0, 2)2\pi/c \pm \vec{k}_0$, at which only two peaks are expected, either because branches are crossing in pairs or because two branches have zero energy. Thus by beginning at values of \vec{Q} where relatively few peaks are expected and by utilizing qualitative theoretical intensity considerations, one can unambiguously map out the dispersion relations for $\pm\vec{q}$ in spite of the rather complicated energy spectrum generally observed for the scattered neutrons. The results obtained for several typical constant- \vec{Q} experiments are shown in Fig. 1. Note that $\vec{k}_0 = (0, 0, 0.24)2\pi/c$. Numerous scans similar to those included in Fig. 1 were used to construct the dispersion relations for $\hbar\omega(-\vec{q})$ and $\hbar\omega(+\vec{q})$ shown in Fig. 2.

The interpretation of these data has been based on the model Hamiltonian which generally is used for the interpretation of the magnetic properties

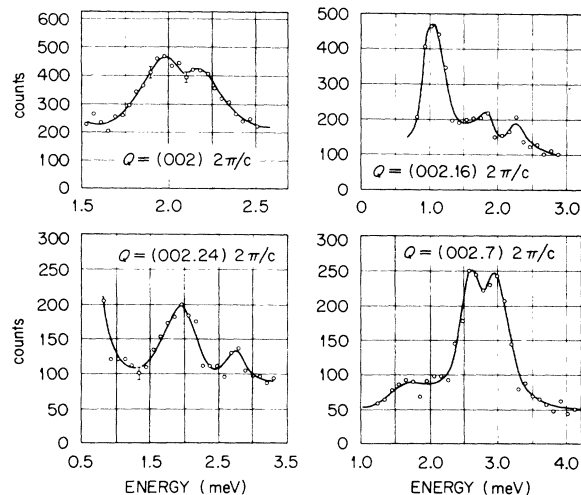


FIG. 1. Neutron intensities observed in several constant- \vec{Q} experiments for erbium.

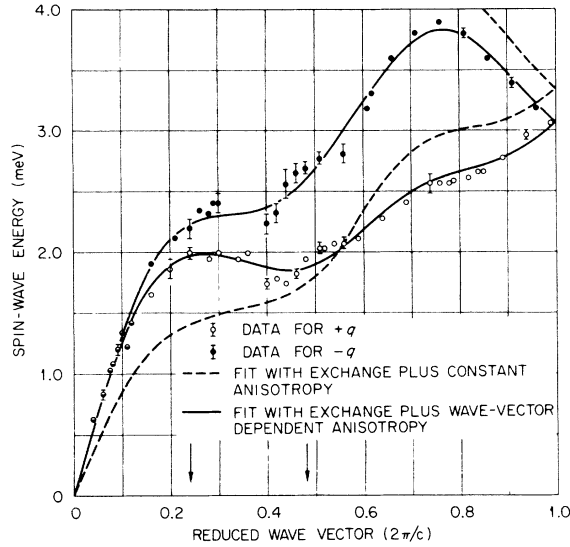


FIG. 2. Spin-wave energies for erbium metal at 4.5°K.

of the heavy rare-earth metals,⁵⁻⁷ and which leads to the following expression for the spin-wave energies along the c axis, in the conical phase, at 0°K:

$$\hbar\omega(\vec{q})/S = \frac{1}{2}[J(\vec{k}_0 + \vec{q}) - J(\vec{k}_0 - \vec{q})]\cos\theta + (F_1 F_2)^{1/2}, \quad (1)$$

where

$$F_1 = J(\vec{k}_0) - \frac{1}{2}J(\vec{k}_0 + \vec{q}) - \frac{1}{2}J(\vec{k}_0 - \vec{q}), \quad (2)$$

$$F_2 = F_1 \cos^2\theta + \{J(\vec{k}_0) - J(\vec{q}) + 2[L - K(\vec{q})]\} \sin^2\theta. \quad (3)$$

Here S is the total angular momentum on each ion; $J(\vec{q})$ is the Fourier transform of the exchange interaction; $L - K(q)$ is a combination of the axial crystal-field anisotropy parameters and an anisotropic exchange interaction; and θ is the angle (28.5° for Er⁸) between the magnetic moment on each ion and the c axis.

The first term on the right side of Eq. (1) changes sign when \vec{q} is changed to $-\vec{q}$. Thus, the difference

$$\Delta(\vec{q}) \equiv \hbar\omega(-\vec{q}) - \hbar\omega(+\vec{q}) = S[J(\vec{k}_0 + \vec{q}) - J(\vec{k}_0 - \vec{q})]\cos\theta \quad (4)$$

obtained experimentally permits a rather direct evaluation of the isotropic exchange interaction. As is customary we assume that the existence of the helical component of the conical structure requires $J(\vec{q})$ to have a maximum for $\vec{q} = \vec{k}_0$. Therefore $\hbar\omega(-2k_0) > \hbar\omega(+2k_0)$ since $J(\vec{k}_0) > J(3\vec{k}_0)$ and

TABLE I. Exchange and anisotropy constants for erbium metal.

$$L - K(\vec{q}) = K_0 + 2 \sum_{l=1}^n K_l \cos \zeta l \pi, \quad |\vec{q}| = 2\pi \zeta / c.$$

Exchange constants J_l (meV)	
J_1	0.050 ± 0.002
J_2	-0.019 ± 0.001
J_3	0.007 ± 0.001
J_4	-0.007 ± 0.0004
J_5	0.0003 ± 0.0004
J_6	0.0004 ± 0.0006
J_7	0.0010 ± 0.0003
Anisotropy constants K_l (meV)	
K_0	4.00
K_1	1.12
K_2	0.36

$J(\vec{q}) = J(-\vec{q})$; and on the basis of the neutron intensity considerations mentioned above, we conclude that $\Delta(\vec{q}) \geq 0$ for all \vec{q} at which measurements were obtained.

In the present work the exchange interaction along the c direction was evaluated by first expressing $J(\vec{q})$ in terms of a cosine series with interplanar exchange constants as coefficients, viz.,

$$J(\vec{q}) = 2 \sum_{l=1}^n J_l \cos(\zeta l \pi), \quad (5)$$

with $|\vec{q}| = 2\pi \zeta / c$ and $\zeta \leq 1$.

With the exchange interaction so expressed, a linear least-squares fitting of Eq. (4) to the experimental results for $\Delta(\vec{q})$ was carried out to obtain the J_l constants, and the results are given in Table I together with the uncertainties that were calculated in conjunction with the fitting analysis. Seven constants give a reasonable fit to the results. However, even with seven constants certain structure observed in the $\hbar\omega(-\vec{q})$ branch between $\zeta = 0.4$ and 0.65 cannot be reproduced. Nevertheless, assuming the validity of Eq. (1), the procedure just described yields a relatively unambiguous (e.g., $\sim \pm 10\%$ for $\zeta = 1$) determination of the magnitude of the exchange interaction, and it provides a qualitative indication of the \vec{q} dependence of $J(\vec{q})$.

The $J(\vec{q})$ obtained for Er is compared in Fig. 3 with results obtained recently for Dy in its helical magnetic phase.⁹ To make this comparison,

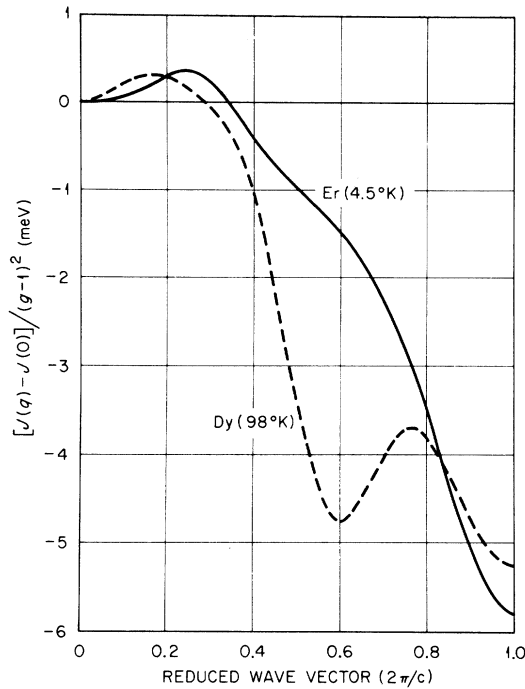


FIG. 3. Comparison of the exchange interaction for Er and Dy.

we have scaled the exchange for both metals by $(g-1)^2$, where g is the Landé factor, since the interaction is believed to be an effective one between spins, whereas the Hamiltonian on which Eq. (1) is based was expressed in terms of the total (spin + orbital) angular momentum of each ion. While the overall magnitude obtained for the exchange interaction of Er is reasonably consistent with that expected, the shape of $J(\vec{q})$ for Er is quite different from the results for Dy and that reported for the other rare-earth metals.^{2, 9-11}

After the determination of the exchange constants J_i from the analysis of $\Delta(\vec{q})$, the anisotropy term $L - K(q)$ in Eq. (1) was then evaluated by a fitting analysis of both the $\hbar\omega(+\vec{q})$ and $\hbar\omega(-\vec{q})$ branches. For this fitting the exchange contributions to F_1 and F_2 in Eq. (1) were computed from the J_i constants given in Table I. Surprisingly the experimental results cannot be described at all with a constant (i.e., \vec{q} -independent) anisotropy, as would result if only the crystal-field contributions to the anisotropy were significant. The very poor fit to the $\hbar\omega(+\vec{q})$ branch that is obtained with a single anisotropy parameter is illustrated by the dashed line in Fig. 2. An improved fit to $\hbar\omega(+\vec{q})$ can be achieved by varying the seven J_i constants as well as the anisotropy constant in the fitting calculation; however, the fit to the $\hbar\omega(-\vec{q})$ branch and, hence, to $\Delta(\vec{q})$ is

then very poor. To obtain a satisfactory fit to these measurements such as that illustrated by the full line in Fig. 2, at least three anisotropy parameters, defined in Table I, are required. The constant K_0 deduced here is approximately an order of magnitude larger than the few tenths of a meV that is estimated from the crystal-field calculations of Kasuya.¹² Also the K_1 and K_2 constants, which presumably describe an anisotropic exchange interaction, are an order of magnitude larger than the isotropic exchange constants.

To our knowledge, these results represent the first definite experimental evidence that for a rare-earth metal either the anisotropic exchange interaction can be very large or the usual model Hamiltonian is not adequate (or both). Additional evidence concerning the inappropriateness of the present model, granting the possibility of such a large anisotropy interaction, is obtained from an inspection of the neutron intensities observed for the branches of the spin-wave dispersion relation. The qualitative (i.e., strong versus weak) relative intensities that are predicted theoretically for the multiple peaks observed at a given Q are in good agreement with our measurements, and as mentioned above, this information enables one to sort the data into the $\hbar\omega(+\vec{q})$ and $\hbar\omega(-\vec{q})$ branches. On the other hand some difficulty was experienced in obtaining a semiquantitative agreement between experiment and theory when the exchange and anisotropy parameters which provide a good fit to the magnon energies are used in the intensity calculation. For example, with the parameters given in Table I, we calculate a 2:1 intensity ratio for the two peaks observed at $\vec{Q} = (0, 0, 2.24)2\pi/c$ shown in Fig. 1, whereas the observed ratio is nearer 4:1. Also at slightly larger \vec{Q} the weaker peak essentially disappears into the background while the intensity of the stronger peak remains high, implying an experimental intensity ratio in excess of 10:1, whereas the largest ratio calculated is only 3.5:1.

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Hybrid Model for Pre-Equilibrium Decay in Nuclear Reactions*

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Ideas from Griffin's exciton model are combined with those from the nucleon-nucleon scattering approach to nuclear transition times to provide a simple closed-form expression for predicting pre-equilibrium decay phenomena, including variation of pre-equilibrium emission with target mass, excitation energy, and initial particle and hole numbers. Time estimates for pre-equilibrium emission are given at several excitations.

The question of the attainment of equilibrium in medium-energy nuclear reactions is very old and of great importance to statistical models. Recent approaches to models which may in time provide answers to this question have been the very simple exciton model proposed by Griffin¹ and the more elaborate master-equation approach due to Miller and co-workers.^{2,3} We propose a model which in some ways provides a marriage between the simple exciton model and the more elaborate model of Refs. 2 and 3. The result maintains the simple closed-form simplicity of the model of Griffin and may be used to predict such properties as the variation of fraction pre-equilibrium emission (hereafter referred to as *f_{pe}*) as a function of excitation energy, compound-state mass number, and initial particle and hole numbers. This may be done either on an *a priori* basis, or by a normalization of a "mean free path" (*mfp*) constant at one excitation. Statements may also be made as to the length of time during which pre-equilibrium emission occurs, as well as to other lifetime averages.

As in high-energy cascade calculations and in Refs. 1-6, it will be assumed that a reaction proceeds through a series of particle-particle or particle-hole interactions, in which the total particle and hole numbers characterizing the nuclear state may either increase by two, decrease by two, or remain unchanged. As in earlier work, we assume that the transitions in which

the particle and hole (*p-h*) or exciton (*n*) number increases by two dominate in the early stages of the equilibration process. As in Griffin's model, we assume that the intermediate states are characterized by appropriate level density formulas and that all levels may be populated with equal *a priori* probability (within limitations of energy conservation and the Pauli principle) during the equilibration process, at least insofar as they "count" the number of ways different transitions may lead to the various final states. However, as in the model of Ref. 2 we recognize that whether or not one particle in a virtual level is emitted into the continuum or undergoes a transition to a more complicated (*n* + 2)-exciton state depends upon the appropriate particle decay rates $\lambda_c(\epsilon)$ and $\lambda_{n+2}(\epsilon)$ for the particle of interest and not for the average over the *n*-exciton state. The level densities as used therefore represent a bookkeeping procedure for the number of ways excitons from a simple state may populate a given energy range in a more complex state, averaged over time, not at a specific time. As in earlier treatments of the exciton model the total particle-emission probability in a given channel energy range $P_x(\epsilon)d\epsilon$ is given as a sum over the contributions of the intermediate states, although here this has significance as a statistical bookkeeping operation rather than on an absolute time basis. The sum is taken from some initial number of excitons n_0 to the equilibrium number