Dynamics of degenerate local excitations in cubic metals

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The problem of an isolated ion with a total angular momentum J in a cubic metal is analyzed by the technique of Abrikosov pseudofermions. A system of generalized kinetic equations is constructed. The particular example $J=5/2$ is studied in the case in which the ground state is the quartet Γ_8 (CeB_6). The central peak in the susceptibility can be described by a sum of two Lorentzians, one of which is narrower and higher than the other. This multimode behavior is shown to be a general phenomenon when a degenerate multiplet is not a doublet and does not constitute all the states of the system.

Solids exhibit a very large number of local excitations with internal degrees of freedom. Many of these excitations are associated with ions of rare-earth elements and actinides with a total angular momentum J. In the crystal field, the corresponding $(2J+1)$-fold multiplet splits into several sublevels. Transitions between these sublevels occur because of an interaction of this multiplet with other degrees of freedom of the crystal (conduction electrons, phonons, etc.), and also because of an exchange interaction between neighboring ions. Associated effects are currently being studied actively. In the present letter we wish to discuss the properties of an isolated ion with a degenerate ground state in a cubic metal. We are particularly interested in the case in which this ground state is not a Kramers doublet. A physical realization of such a system is the compound CeB_6. In this case we have $J=5/2$, the ground state of the Ce^{3+} ion is the quartet Γ_8, the excited state is the doublet Γ_7, and the distance between these states is 46 meV (Ref. 1). At $T_Q=3.1$ K, however, there is a transition to a quadrupole state in the subsystem of Ce^{3+} ions.[2] The cerium ions can thus be assumed to be independent at $T \gg T_Q$. We will also be ignoring consequences of the Kondo effect. According to Kunii,[3] the Kondo temperature in CeB_6 is lower than 10 K. For CeB_6, our theory thus applies at $T \gg 10$ K.

The problem of an isolated rare-earth ion in a cubic metal has been studied theoretically by Becker[4] (see also the review by Fulde and Loewenhaupt[5]) by the method of Zwanzig-Mori memory functions. In particular, the example of the Ce^{3+} ion was studied in the case in which the ground state is the Γ_7 doublet. In the present letter we use the simpler and more graphic method of pseudofermions, as proposed by Abrikosov.[6] The results of Refs. 4 and 5 on the Γ_7 ground state are basically the same as our own. If the lower level is instead the Γ_8 quartet, the dynamic susceptibility is described by a sum of two Lorentzians; i.e., the central peak has a two-mode structure.

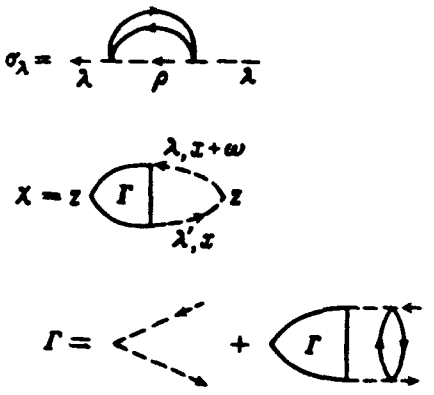

FIG. 1.

This multimode behavior is characteristic of the central peak when the degeneracy of the ground state is higher than a Kramers doublet.

The Hamiltonian of this system is

$$H=H_{\rm el}+\sum_{\lambda}(\epsilon_\lambda-\mu)a_\lambda^+a_\lambda+(1/2)(g_J-1)J_{\rm ex}\sum(a_\rho^+\mathbf{J}_{\rho\lambda}a_\lambda)\cdot(\psi_\varphi^+\boldsymbol{\sigma}_{\varphi\nu}\psi_\nu), \qquad (1)$$

where $H_{\rm el}$ is the Hamiltonian of the conduction electrons, a_λ is a pseudofermion operator which annihilates a state λ of the multiplet with an energy ϵ_λ, μ is the chemical potential, g_J is the Landé g-factor, and ψ_φ is an operator which annihilates an electron at an ion site. To ensure that one and only one of the states λ is occupied, we need to take the limit[6] $\mu\to-\infty$. We will calculate the susceptibility of an isolated ion with a total angular momentum **J**. In a cubic crystal this susceptibility is given by

$$\chi(\omega)=i\int_0^\infty dt e^{i\omega t}\langle[J_z(t),J_z(0)]\rangle. \qquad (2)$$

A Feynman-diagram series for χ in lower-order perturbation theory is shown in Fig. 1. The dashed and straight lines correspond to pseudofermions and electrons.

The real part of the pseudofermion eigenenergy $\sigma_\lambda(\epsilon_\lambda)$ corresponds to a shift of the level λ. This real part depends weakly on the temperature and can be incorporated in the definition of ϵ_λ. The quantity $-{\rm Im}\sigma_\lambda(\epsilon_\lambda)=\gamma_\lambda$ determines the width of the λ level.

Continuing from discrete imaginary frequencies to the real axis, we easily find the formula[7-9]

$$\chi(\omega)=\frac{1}{2\pi iN}\int dx e^{-x/T}[g(x)\Gamma(x,x+\omega)g(x+\omega)-g^*(x)\Gamma(x,x+\omega)g(x+\omega)$$
$$+g^*(x-\omega)\Gamma(x-\omega,x)g(x)-g^*(x-\omega)\Gamma(x-\omega,x)g^*(x)]J_z. \qquad (3)$$

Here g and Γ are the pseudofermion Green's function and the vertex part; the state labels have been omitted; $g^*(x)=g(x-i\delta)$; and the imaginary parts of the arguments of the function Γ are the same as for the corresponding g-functions. The derivation of

(3) incorporated the circumstance that Γ is an analytic function of its two independent variables with cuts along the real axis.[10] The normalization factor N in (3) is

$$N=\sum_{\lambda}\langle a_{\lambda}^{+}a_{\lambda}\rangle=\sum_{\lambda}G_{\lambda}\exp(-\epsilon_{\lambda}/T), \tag{4}$$

where G_{λ} is the degree of degeneracy of level λ. In this expression, as in (3), we have taken the limit $\mu\to-\infty$. Making use of the analytic properties of the vertex Γ and the circumstance that the singularities of the g-function lie at $+\infty$, we easily find an equation for Γ for real variables from the condition $\mu\to-\infty$ (Refs. 8 and 9). If the imaginary parts of the two arguments are identical, the integral term is small, and the corresponding vertices are the same as J_z. If the imaginary parts of the arguments are instead different, the poles of the Green's functions lie on different sides of the real axis. If damping is ignored, the integral diverges. As a result, in the case $\gamma_{\lambda}\ll T$ we find the system of equations

$$\Gamma_{\lambda'\lambda}(x,x+\omega)=J^{z}_{\lambda'\lambda}+2ig^{2}\mathbf{J}_{\lambda'\nu}\frac{\epsilon_{\nu\lambda'}N(\epsilon_{\nu\lambda'})\Gamma_{\nu\mu}(\epsilon_{\nu},\epsilon_{\nu}+\omega)}{\omega+\epsilon_{\nu\mu}+i\gamma_{\mu}+i\gamma_{\nu}}\mathbf{J}_{\mu\lambda}, \tag{5}$$

where $\epsilon_{\nu\lambda'}=\epsilon_{\nu}-\epsilon_{\lambda'}$; N is the Planck function; a summation is carried out over μ and ν; and we have $g^{2}=(\pi/2)[(g_{J}-1)J_{ex}N_{0}]^{2}$, where N_{0} is the density of electron states at the Fermi level. This expression is valid in the case $g^{2}\ll 1$. Equations (5) constitute a generalized kinetic equation which describes transitions between levels (off-diagonal $\Gamma_{\lambda'\lambda}$) and fluctuations in their populations ($\Gamma_{\lambda\lambda}$) caused by a perturbation proportional to J_z.

As an example we consider an ion with $J=5/2$. The wave functions are[4,5]

$$\psi_{1\pm}=a|\pm 5/2\rangle-b|\mp 3/2\rangle;(\Gamma_{7}),$$

$$\psi_{2\pm}=b|\pm 5/2\rangle+a|\mp 3/2\rangle,\ \psi_{3\pm}=|\pm 1/2\rangle;(\Gamma_{8}), \tag{6}$$

where $a=(1/6)^{1/2}$ and $b=(5/6)^{1/2}$. The interaction of the total angular momentum $\mathbf{J}$ with electrons does not commute with the Hamiltonian of the crystal field. As a result, terms appear in (5) which mix fluctuations of the populations of the components $\psi_{2\pm}$ and $\psi_{3\mp}$ of the Γ_8 quartet. This mixing stems from the fact that the matrix elements of the operators $J_{\pm}$ between the states $\psi_{2\pm}$ and $\psi_{3\mp}$ are not zero. This point can be ignored if the ground state is the Γ_7 doublet and if the distance between levels satisfies $\epsilon\gg T$. In the case of a Γ_8 ground state, and under the condition $\epsilon\gg T$, on the other hand, the effect of Γ_7 can be ignored in a first approximation, and under the condition $\omega\ll\epsilon$ the dynamic susceptibility is determined by a relaxation of the populations of the states of the quartet. In this case a mixing of the states $\psi_{2\pm}$ and $\psi_{3\mp}$ plays a governing role. As a result, using $\Gamma_{2(3)+,2(3)+}=-\Gamma_{2(3)-,2(3)-}=\Gamma_{2(3)}$, we can put system (5) in the form

$$(\omega+62ig^{2}T/9)\Gamma_{2}+4ig^{2}T\Gamma_{3}/3=11(\omega+2i\gamma_{2})/6,$$

$$4ig^{2}T\Gamma_{2}/3+(\omega+58ig^{2}T/3)\Gamma_{3}=(\omega+2i\gamma_{2})/2, \tag{7}$$

where $\gamma_2 = 65g^2T/12 = -\mathrm{Im}\sigma_{2(3)}(0)$ is the damping of the levels of the quartet. Solving this system of equations, and substituting the result into (3), we find the following expression for the susceptibility under the condition $\omega \ll \epsilon$:

$$\chi_{\mathrm{Rel}}(\omega) = \frac{1}{T}\left(\frac{2i\gamma_+ Z_+}{\omega + 2i\gamma_+} + \frac{2i\gamma_- Z_-}{\omega + 2i\gamma_-}\right), \tag{8}$$

where

$$Z_\pm = \pm\left[\frac{359\times 11}{9}\left(\delta_\pm - \frac{313}{33} + 109(\delta_\pm - 1)\right)\right][72(\delta_+ - \delta_-)\delta_\pm]^{-1}, \tag{9}$$

and

$$\gamma_\pm = g^2 T\delta_\pm, \ \delta_\pm = [59 \pm (820)^{1/2}]/9. \tag{10}$$

The central peak, which describes the relaxation of the population of the quartet components, thus has a two-mode structure. In terms of numerical values we have $\delta_+ = 9.74$, $\delta_- = 3.37$, $(\delta_-/\delta_+) = 0.347$, $Z_+ = 0.238$, $Z_- = 1.57$, and $Z_-/Z_+ = 6.58$. We see that nearly all the intensity is in the minus mode, which relaxes three times as fast as the plus mode.

The resonant contribution to $\chi(\omega)$ is of the same form as in Refs. 4 and 5:

$$\chi_{\mathrm{Res}}(\omega) = -\frac{20(1 - e^{-\epsilon/T})}{9(2 + e^{-\epsilon/T})(\omega - \epsilon + 1\gamma_{21})}, \tag{11}$$

where $\gamma_{21} = g^2(10/3)\{\epsilon[3N(\epsilon) + 2] + 35T/12\}$. This expression is also valid at $T \sim \epsilon$. Expressions (8) and (11) saturate the standard sum rule:

$$\langle J_z^2 \rangle = -(1/\pi)\int d\omega(\mathrm{Im}\,\chi_{\mathrm{Rel}} + \mathrm{Im}\,\chi_{\mathrm{Res}})N(-\omega). \tag{12}$$

If the ground state is the Γ_7 doublet, the results at $T < \epsilon$ are the same as in Refs. 4 and 5.

We conclude with the following comments.

1. The derivation of (7)-(10) was restricted by the condition $\epsilon \ll T$. However, the initial kinetic equations, (5), are valid for an arbitrary relation between ϵ and T. The derivation of those equations was limited by the conditions $g^2 \ll 1$, $\gamma_\rho < T$, and $g^2 \ln[E_F/\min(\epsilon_\lambda T)] < 1$. The latter condition means that we can ignore corrections for the Kondo effect.

2. Equations like (5) can be written for the cases of an interaction with phonons or spin waves. In those cases, however, the amplitudes for transitions between degenerate states are zero, since the density of states of the corresponding excitations tends toward zero more rapidly than ω at small values of ω. It is thus necessary to take into account two-quantum Raman processes, as was done in Ref. 8 in a study of the interaction of a degenerate center with phonons.

3. We have found that the central peak has a two-mode structure because of a mixing of fluctuations of the populations of different components of the Γ_8 quartet. If we had been discussing the quadrupole susceptibility instead of the ordinary suscep-

tibility, we would have found a two-mode structure again, but with different relaxation times, corresponding to other zeros of the fourth-order determinate which determines the properties of the solution of Eq. (5) in the case of four diagonal vertices $\Gamma_{\lambda\lambda}$.

In general, the following conditions must hold if the central peak is to have a multimode structure: 1) The degree of degeneracy of the ground state must be greater than 2. 2) The system must have at least one excited state in addition to the degenerate state which we have been discussing. Otherwise,[10] we would have $\Gamma \sim J_z$, and we should find the single-mode behavior immediately from (5). This assertion is clearly illustrated by the example of an ion with $J=3/2$. In this case, as above, it is simple to find two modes: $2\gamma_+ = 12g^2T$ and $2\gamma_- = 2g^2T$. However, Eq. (5) is constructed in such a way that the mode $2\gamma_+$ disappears from the solution, and we have

$$\Gamma = J_z(\omega + 2i\gamma)/(\omega + 2ig^2T), \tag{13}$$

where $\gamma = (15/4)g^2T$.

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