MODIFIED SPIN SPLITTING OF LANDAU LEVELS OBSERVED IN A KONDO SYSTEM

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The de Haas–van Alphen effect has been studied in the Kondo system Cu:Cr in magnetic fields up to 55 kG at 1.0 K. In alloys containing ~30 ppm Cr "beat" minima and shifts of the spin-splitting zeros are observed for several orbits. These observations are explained by supposing the normal spin splitting of the Landau levels to be modified by an antiferromagnetic exchange term, so that the effective g factor becomes $g - \Delta\epsilon_{ex}/\mu_B H$.

Some time ago we reported[1] the observation of a sharp minimum in the amplitude of $\langle 111\rangle$ "belly" de Haas–van Alphen (dH–vA) oscillations in very dilute (~30 ppm) Cu:Cr alloys. The effect had the appearance of a "beat" minimum, but could not be ascribed to sample imperfection, so we postulated at the time the existence of an electron-scattering anomaly. It now appears probable that we are indeed observing a form of beat and that this is fundamental in origin.

The frequencies observed in the dH–vA effect can be related to an extremal area of the Fermi surface and are associated with the spacing of the Landau levels for that cross section. The amplitude of the oscillations is a complex function of magnetic field H and temperature T but is also affected by the electron spin. Each of the Landau levels is split according to the spin direction, introducing into the amplitude a term $\cos(\pi g m^*/2m_0)$, where the factor g is usually but not always 2 and m^*/m_0 is the cyclotron mass in units of free-electron mass. When the spin splitting is exactly half the spacing of the Landau levels, the amplitude of the fundamental drops to zero; this corresponds to gm^*/m_0 taking the value 1, 3, etc.

In ferromagnetic systems such as iron,[2] nickel,[3] or palladium with a small concentration of cobalt,[4] an exchange interaction splits the Fermi surface into separate spin-up and spin-down surfaces (with corresponding extremal areas) and the spin-splitting factor is suppressed. This feature is particularly striking in the experiments of Hornfeldt, Ketterson, and Windmiller,[4] where small concentrations of cobalt added to palladium suppressed the spin-splitting zeros, and the nesting spin-up and spin-down Fermi surfaces caused a beat in the dH–vA oscillations. This experiment suggested that we reinterpret our data for Cu:Cr in terms of a beat induced by the localized moment. We used for new experiments a single crystal of copper doped with 35 ppm (nominal) chromium. The resistance ratio of the specimen (room temperature to 4.2 K) was 42, giving a value of 34 ppm using the resistivity data of Daybell and Steyert.[5]

The amplitude of the dH–vA oscillations in susceptibility is conventionally given by

$$A(H,T,x) = (M_0 T/H^{1/2})\exp[-\alpha m^*(T+x)/H],$$

where α is a constant, M_0 and m^* are characteristics of the orbit, and x is the Dingle or scattering temperature. Shown in Fig. 1 are data for $\langle 111\rangle$ belly oscillations in the 34-ppm Cr in Cu alloy and a curve calculated as

$$A(H,T,x)|\cos[\pi(\Delta F/H+\gamma)]|,$$

where M_0 and x are fitted to amplitudes at extreme field values. γ is initially assumed to be zero, and the beat frequency ΔF is then related to the field H_{min} at which the minimum occurs by $\Delta F/H_{min} = \frac{1}{2}$. As will be seen later, there is

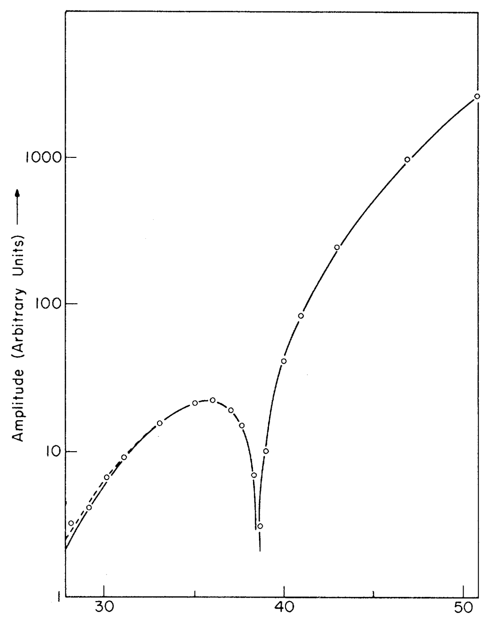

FIG. 1. Experimentally determined amplitudes for the $\langle 111\rangle$ belly in Cu+34 ppm Cr at ~1 K, and fitted curves of the form $A(H,T,x)|\cos(\pi\Delta F/H)|$ (solid and dashed) and $A'(H,T,x)|\cos[(\pi/2)(gm^*/m_0 - 2\Delta F'/H)]|$ (solid line).

some flexibility in the value of γ when making a fit to a single minimum, but the rather good agreement obtained suggests strongly that the dip in the amplitude does arise from a beat. Similar behavior, with different values of $H_{\rm min}$ (see Table I), was observed in all other orbits with the exception of the "neck" orbit when the field direction was very close to ⟨111⟩. In this case no sign of a beat could be found even though we extended the search from a maximum of ~55 kG to below 10 kG, where all signal disappeared.

An obvious explanation for these beats which must be considered is the possibility of specimen imperfection. If two portions of the specimen are at slightly different orientations they will produce dH–vA frequencies which differ slightly and which beat together. This explanation can be eliminated for a number of reasons: The specimen was chosen carefully for crystalline perfection, the phenomenon is reproducible from specimen to specimen (cut from the same ingot), and the position and depth of the minimum is highly temperature dependent.[1]

If we ascribe these beats to an exchange term splitting the Fermi surface into separate spin-up and spin-down surfaces, we would expect to find the normal spin splitting of the Landau levels suppressed so that the usual vanishing of the dH–vA amplitude when $gm^*/m_0 = 1$, 3, etc. would not occur. In Fig. 2 we compare parts of the rotation diagram (dH–vA oscillations obtained by rotating the crystal in a constant magnetic field) for pure Cu, where spin-splitting zeros

Table I. Positions of minima for various orbits. The values of m^*/m_0 at the symmetry directions are experimental, those away from the symmetry directions are derived using the data of Koch, Stradling, and Kip.[a] The value of $\Delta\epsilon/\mu_B$ is calculated by taking $g=2$ and assuming $(m^*/m_0)(g-\Delta\epsilon/\mu_B H_{\rm min})$ to be 1 except for the cases marked (†) where the value −1 provides the upper limit shown.

Orbit	Angle from <100> (degrees)	m^*/m_o	$H_{\rm min}$ (kilogauss)	$\Delta\epsilon/\mu_B$ (kilogauss)
Belly	0	1.35	30.3	38.1
Rosette	0	1.30	32.3	39.7
Belly	16.2	1.32	34.7	43.0
Belly	54.75	1.36	38.6	48.5
Neck	54.75	0.46	<5	<21†
Neck	70.5	0.54	<10 or >100	<38† and >15
Neck	76.3_5	0.70_8	54.15	31.9
Neck	76.9_5	0.72_8	50.85	32.1
Neck	77.5_5	0.75_2	47.6	32.1
Dogsbone	90	1.29	27.6	33.6

[a]J. F. Koch, R. A. Stradling, and A. F. Kip, Phys. Rev. 133, A240 (1964).

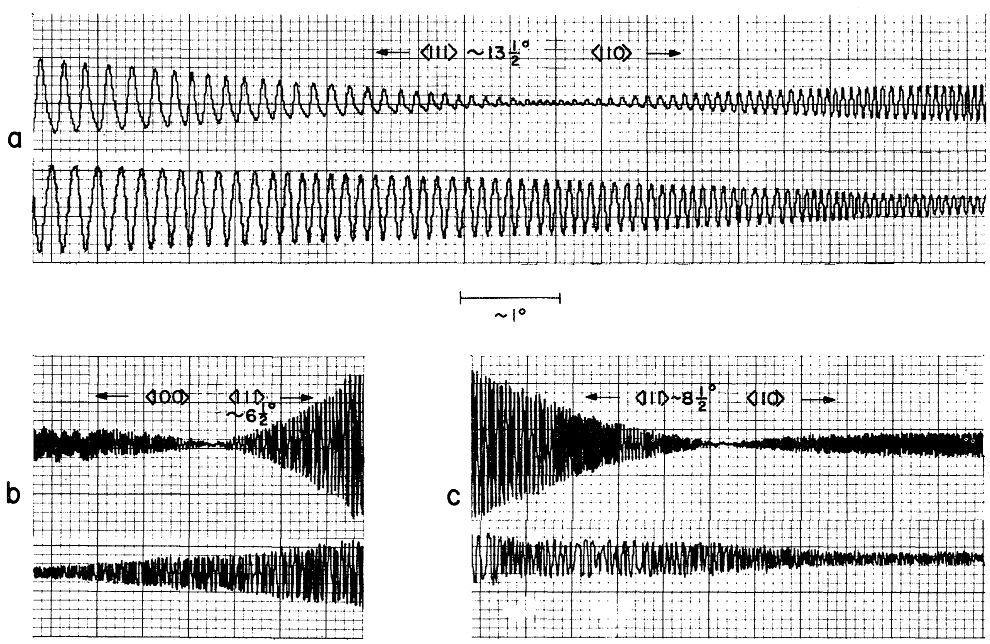

FIG. 2. Rotation diagrams in the (110) plane at 50.85 kG and ~1 K for pure Cu (upper traces) and Cu+34 ppm Cr (lower traces). (a) Neck oscillations, ~63° to 73°; (b) belly, ~46.5° to 50°; (c) belly, ~61° to 66.5°; all angles measured from ⟨100⟩. The small horizontal steps in the oscillations derive from the windings on the potentiometer used to indicate angle, and hence are spurious.

occur, with the corresponding regions in the alloy, where the spin-splitting zeros have been eliminated as expected. However, when the neck oscillations in the alloy were followed on a rotation diagram to an angle some 8° further from the symmetry direction, we observed what appeared to be a new spin-splitting zero. Further investigation showed that the angular position of this minimum varied with magnetic field, so it could be studied either as a function of angle at constant field, when it looked like a normal spin-splitting zero, or as a function of field at constant angle, when it looked like a beat. The results of this examination are shown in Table I.

We explain these results in the neck frequency by proposing that the conventional spin-splitting term, $\cos[(\pi/2)gm^*/m_0]$, be replaced be a term $\cos[(\pi/2)(gm^*/m_0-2\Delta F/H)]$, where ΔF is a beat frequency. The negative sign is necessary to fit the observed decrease of $H_{\min}$ with increasing effective mass. We relate the beat frequency to an energy term $\Delta\epsilon$ by using the Onsager relation $\Delta A=(\hbar/2\pi e)\Delta F$ and the effective mass $m^*=(\hbar^2/2\pi)(dA/d\epsilon)$. The modified spin-splitting term then becomes $\cos[(\pi/2)(m^*/m_0)(g-\Delta\epsilon/\mu_B H)]$. Furthermore, we postulate that the parameter $\Delta\epsilon$ is only weakly dependent on orientation save for orbits near the zone boundary where it becomes small for reasons that will become apparent below. This modified spin-splitting term not only explains the neck behavior but also can account for the beats observed in other orbits. It is apparent from Fig. 1 that the amplitude behavior is insensitive to the exact phasing of the beat, provided it is fitted at the beat minimum; the two expressions shown $\{\cos[(\pi/2)(gm^*/m_0-2\Delta F/H)]$ and $\cos(\pi\Delta F/H)\}$ can be distinguished only in regions which are experimentally inaccessible. The values of $\Delta\epsilon/\mu_B$ derived from our experimental results are shown in Table I.

We justify the formula postulated above by assuming the Landau levels to be split according to spin orientation by two terms: the ordinary spin splitting given by $\pm g\mu_B H/2$, and a constant energy splitting, attributed to an exchange term $\pm\Delta\epsilon_{ex}/2$. The first term gives the conventional factor derived by Dingle[6] of $\cos[(\pi/2)gm^*/m_0]$. If the exchange is ferromagnetic a similar treatment gives a factor $\cos[(\pi/2)(m^*/m_0)(g+\Delta\epsilon_{ex}/\mu_B H)]$; if antiferromagnetic, the factor $\cos[(\pi/2)(m^*/m_0)(g-\Delta\epsilon_{ex}/\mu_B H)]$ that we postulate above. We note that the actual behavior observed indicates unambiguously an antiferromagnetic exchange term in our Kondo system.

It is natural to identify this antiferromagnetic exchange splitting with the s-d exchange term introduced by Kondo.[7] The exchange Hamiltonian can be written

$$H_{ex}=-J\mathbf{S}\cdot\sigma\Omega,\ \ J<0,$$

where the impurity has a spin S, the conduction-electron spin density is σ, Ω is the atomic volume, and J the exchange parameter. If the impurity concentration is c we expect

$$\Delta\epsilon_{ex}\sim cSJ.$$

Taking $S=\frac{3}{2}$ (Daybell, Pratt, and Steyert[8]) and $\Delta\epsilon_{ex}$ from the $\langle 111\rangle$ belly orbit (Table I), we obtain for our 34-ppm alloy a value of J of order 5.5 eV which is comparable with the value of 1.8 eV derived from the expression[9]

$$T_K\sim T_F\exp[-1/N(\epsilon)|J|]$$

using a Kondo temperature[8] T_K of 1.0 K, and free-electron values for the Fermi temperature T_F, and the density of states $N(\epsilon)$ per atom for one spin index.

The parameter J (and thus $\Delta\epsilon_{ex}$) will be anisotropic; in particular near the neck, where the wave function is p like, we expect J (the s-d interaction constant) to be small if not zero. This behavior is reflected in the $\Delta\epsilon$ values listed in Table I which show a decrease for orbits that approach the $\langle 111\rangle$ zone boundary such as the dogsbone and, in particular, the neck at $\langle 111\rangle$.

This simple model suggests that $\Delta\epsilon_{ex}$ (and therefore $H_{\min}$) should vary linearly with concentration. Our earlier experiments[1] and additional experiments in the range 25-35 ppm do in fact suggest that the position of the beat minimum is at least approximately linear with concentration up to 35 ppm, though above this the concentration effects noted in the specific-resistivity results[5] and attributed to impurity-impurity interactions are reflected in the position (and depth) of the minima.

It is interesting to note that at 1.0 K (i.e., near T_K) the beat zero is well defined, being less than 2 % of the maximum amplitude. This suggests that in the region of the Kondo temperature all the conduction electrons are polarized by the localized moment, at least in magnetic fields above 10 kG. As the temperature is raised the beat minimum becomes less well defined, consistent with a decreasing fraction of the electrons being polarized by the localized moment. It is possible that the exchange splitting originates solely from a Ruderman-Kittel-Yosida-type long-range spin polarization about the impurity and is entirely unrelated to the "Kondo" phenomena. However, in that case it becomes difficult to explain the rapid degrading of the beat as the temperature is raised above T_K.

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THEORETICAL ESTIMATES FOR THE DECAY RATES OF THE SPIN FLUCTUATIONS IN $RbMnF_3$†

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We have calculated the decay rates of the spin fluctuations in the hydrodynamic region above T_N for the isotropic antiferromagnetic $RbMnF_3$. Our results agree in magnitude, temperature dependence, and wave-vector dependence with the experimental values reported by Lau et al.

In a recent Letter, Lau et al.[1] have reported measurements of the inelastic scattering of neutrons from $RbMnF_3$ in the vicinity of the Néel point. Among the data presented are values of the decay rates of the spin fluctuations $\vec{S}_{\vec{q}+\vec{K}_0}(t)$ in the hydrodynamic region above the ordering temperature. [Here $\vec{S}_{\vec{K}_0}$ is the staggered spin operator and $q/K_0 \ll 1$.] In this Letter we report calculations of the decay rates which are in substantial agreement with the experimental values in their temperature dependence, wave-vector dependence, and overall magnitude.

Our calculated values are obtained from a self-consistent theory for the damping constants first introduced for isotropic ferromagnets by Bennett and Martin[2] and subsequently extended by Kawasaki to isotropic antiferromagnets[3] and by us to planar ferromagnets.[4] Two desirable features of the self-consistent theory are that it yields results in agreement with the predictions of the dynamic scaling laws[5] and that it involves as parameters the measurable static properties of the system (lattice parameters, exchange constants, susceptibilities, and correlation lengths). Because of this we are able to incorporate considerable empirical information into the theoretical calculation by using experimental values for these parameters wherever they appear.

The basic equations for the isotropic antiferromagnet follow from the analysis given in Ref. 3. The decay rates are obtained from the solutions of coupled nonlinear integral equations involving spin fluctuations with wave vectors in the vicinity of the center of the Brillouin zone and near the point $\vec{K}_0$, the reciprocal lattice vector of the magnetic superlattice. Writing these rates as $\varphi(\vec{q})$ and $\varphi(\vec{q}+\vec{K}_0)$, we have

$$\varphi(\vec{q})=\frac{4}{\chi_0\beta_0 N}\sum_{\vec{p}}{}' \frac{[J(\vec{q}-\vec{p}-\vec{K}_0)-J(\vec{K}_0+\vec{p})]^2}{\varphi(\vec{q}-\vec{p}-\vec{K}_0)+\varphi(\vec{p}+\vec{K}_0)}\chi(\vec{K}_0+\vec{p})\chi(\vec{q}-\vec{p}-\vec{K}_0), \quad (1)$$

$$\varphi(\vec{q}+\vec{K}_0)=\frac{32\chi_0}{\chi(\vec{q}+\vec{K}_0)\beta_0 N}\sum_{\vec{p}}{}' \frac{[J(0)]^2\chi(\vec{K}_0+\vec{p})}{\varphi(\vec{q}-\vec{p})+\varphi(\vec{K}_0+\vec{p})}. \quad (2)$$