Susceptibility and Electron-Spin Relaxation of Fe in Cu below T_K: A NMR Study of ^{63}Cu Satellites

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NMR shifts ΔK and relaxation times T_1 of ^{63}Cu satellite resonances corresponding to nuclei near impurities in *Cu*Fe dilute alloys have been measured for $H \sim 60$ and 30 kG and $T < 20$ K. The $\Delta K(T)$ data confirm that the conduction-electron spin polarization scales with the impurity susceptibility and varies as $1-(T/17)^2$ for $T \ll T_K$. A "Korringa" relation $T_1T(\Delta K)^2$ = const is found, in agreement with recent calculations of the dynamic susceptibility.

Considerable theoretical and experimental effort has been directed towards understanding the static and dynamic properties of Kondo systems.[1] Recent experimental data taken on *Cu*Fe, which has a favorable Kondo temperature ($T_K \sim 30°$K), established that local Mössbauer-effect[2] and macroscopic[3] static susceptibilities (χ_{loc} and χ_m respectively), as well as the excess Knight shift ΔK of nuclei near impurities,[4] all exhibit a $(T+T_K)^{-1}$ temperature dependence. These results imply that the major manifestation of the Kondo effect is variation of χ_{loc} with temperature, rather than any anomalous spin polarization of the electrons below T_K.

The experimental determination of the dynamic susceptibility of the impurity (i.e., the relaxation time τ which characterizes the impurity-spin autocorrelation function) is much less advanced. ESR experiments do not usually yield τ because of the strong bottleneck effect.[5] The impurity nuclear T_1 is directly related to τ. Apart from systems with very high T_K,[1] the experimental requirements in order to observe the NMR are usually very severe.[6] Recent work on copper-based alloys[7] has shown that τ can be deduced from the impurity contribution to T_1^{-1} of ^{63}Cu nuclei. In order to avoid difficulties linked with interactions between impurities and chemical clustering,[8] I have performed transient NMR measurements on the near-neighbor nuclei of the impurities, rather than on the bulk ^{63}Cu nuclei. The experimental results reported here yield the first unambiguous determination of τ at low temperature for *Cu*Fe, and in addition improve by an order of magnitude the accuracy of the NMR shift ΔK for $T \ll T_K$.

Data have been taken at low temperatures ($1.2 < T < 20°$K) at fields of approximately 60 and 30 kG on *Cu*Fe samples with Fe concentrations of 82, 275, 340, 480, and 880 ppm. The use of transient digital recording of spin echoes and signal averaging allowed the detection of all the satellite resonances previously observed by Boyce and Slichter[4] at high temperature.

At fixed T the width of a given satellite increases with concentration, while its position does not change within experimental accuracy.[9] Except for satellite N, which is the nearest to the central line, the present results agree with and improve the accuracy of those of Ref. 4 for $T \lesssim T_K$. In Fig. 1, all available data for $\Delta K/K$ have been plotted versus $(T+27.6)^{-1}$, which has been found to be the best Curie-Weiss fit to the susceptibil-

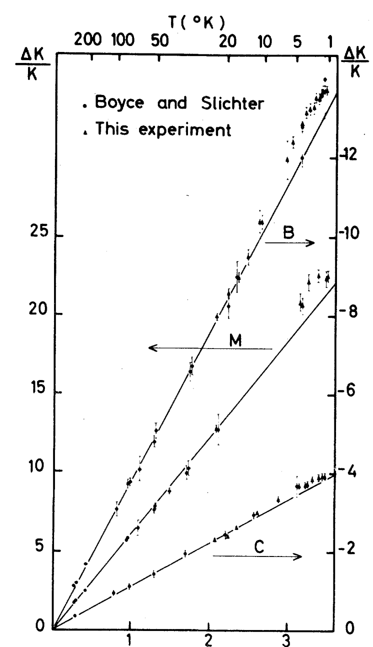

FIG. 1. NMR data for the excess Knight shift ΔK, normalized to the pure copper shift $K = 0.232\%$, plotted versus $(T+27.6)^{-1}$ for the three satellites B, M, and C.

ity data for $T > 15°K$.[2] Although $\Delta K/K$ satisfies the high-T law down to about 15°K, a small relative increase of about 7% of $\Delta K/K$, which was not detected by Boyce and Slichter, is obtained at lower T. Nevertheless the *spatial* dependence of the spin polarization is apparently independent of T, as this increase is the same within experimental accuracy for satellites B, M, and C.

In order to understand whether these deviations correspond to a slight nonlinear dependence of ΔK on $\chi_{\rm loc}$, or to a real T dependence of $\chi_{\rm loc}$, comparison with Mössbauer data is required. Such experiments yield a direct measure of the impurity Knight shift $K(^{57}\mathrm{Fe})$, which is the sum of a spin term $K_d(T) \propto \chi_{\rm loc}(T)$ and of an orbital term $K_{\rm orb}$ usually considered to be independent of T.[1] The shape of $\chi_{\rm loc}(T)$ deduced from the Mössbauer data depends somewhat upon the value taken for $K_{\rm orb}$, as has been shown by various consecutive analyses.[2] As can be seen in Fig. 2, the best NMR data for $\Delta K/K$ agree with the T-dependent contribution to $K(^{57}\mathrm{Fe})$ under the assumption that $K_{\rm orb} = (5 \pm 0.5)\%$. Comparative analysis of $K(^{57}\mathrm{Fe})$ and $\chi_m^{\;2}$ yielded the value $K_{\rm orb} = (5.6 \pm 0.9)\%$. Moreover for $T < 6°K$, for which the exact value of $K_{\rm orb}$ has no real importance, both Mössbauer and satellite NMR measurements clearly show that $\chi_{\rm loc}(T) = \chi_{\rm loc}(0)[1 - (T/\Theta)]^2$, with $\Theta = (17 \pm 1)°K$.

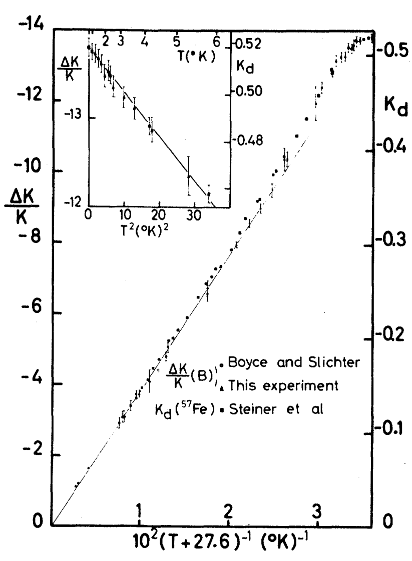

FIG. 2. Comparison between $\Delta K/K$ for satellite B, and the d contribution K_d to $K(^{57}\mathrm{Fe})$. A constant orbital shift $K_{\rm orb} = 0.05$ has been assumed. Typical error bars for K_d, when not given, are the same as for the lowest-T point. In the inset the low-T points are plotted versus T^2.

Such a temperature dependence has already been observed for *Au*V.[10]

In order to measure T_1 of the satellite resonances, appropriate combinations of rf pulses have been used to prepare the nuclear spins in an initial state of uniform spin temperature for all quadrupole transitions of the satellite. The resonance of satellite nuclei (SN) is always superimposed on a background signal associated with other "like" bulk nuclei (LBN). The observed magnetization recovery had correspondingly two distinct time constants $T_1^{\rm SN}$ and $T_1^{\rm LBN}$. A number of experimental and theoretical arguments (which will be published elsewhere) allowed the conclusion that $T_1^{\rm SN}$ is truly related to the spin-lattice relaxation processes, rather than to cross-relaxation processes, at least for shells B and M. Space does not permit a detailed account; however I shall discuss briefly (i) the possibility of spatial diffusion from satellite nuclei to bulk nuclei which have the same resonance frequency, and (ii) spectral diffusion from satellite nuclei to the unshifted copper nuclei. Both processes are expected to decrease with increasing distance $|\Delta K|$ from the main resonance. Since at 1.2°K, the measured values for $T_1^{\rm SN}$, 85±15, 12±1, and 5±1 msec, respectively, for satellites C, B, and M vary in the opposite direction, it can be concluded that such spin-diffusion processes are unimportant, at least for the two outer satellites B and M. This absence of detectable spin-diffusion processes is compatible with the large spin-diffusion barriers for the macroscopic diffusion of

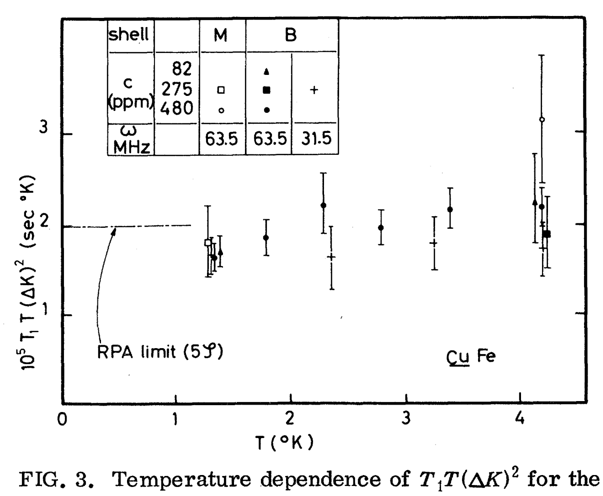

FIG. 3. Temperature dependence of $T_1T(\Delta K)^2$ for the second (M) and third (B) nearest-neighbor shells of the impurity, for various impurity concentrations c and at two frequencies, 63.5 and 31.5 MHz. (RPA is random-phase approximation.)

magnetization evidenced by T_1 measurements on bulk nuclei in copper-based alloys.[7] The data for T_1^{SN} (hereafter T_1) of satellites B and M are summarized in Fig. 3, where the quantity $T_1T(\Delta K)^2$ has been plotted versus T. Within experimental accuracy T_1 was found to be independent of the particular position on the line on which the measurements were performed.

Nuclei with spherical coordinates r, θ, and φ with respect to the impurity are coupled to the Fe magnetization through both an isotropic coupling [$A_s(r)$, Ruderman-Kittel-Kasuya-Yosida (RKKY)] and a much smaller anisotropic one [$A_a(r,\theta,\varphi)$, dipolar and pseudodipolar]. The corresponding relaxation rates for a ^{63}Cu nucleus are given by[7]

$$T_{1s}^{-1} = 2\hbar^{-2}A_s^2(r)k_BT(g\mu_B)^{-2}\,\mathrm{Im}[\chi_T(\omega_n)/\omega_n], \tag{1}$$

$$T_{1a}^{-1} = 2\hbar^{-2}A_a^2(r,\theta,\varphi)k_BT(g\mu_B)^{-2}\,\mathrm{Im}[\chi_L(\omega_n)/\omega_n], \tag{2}$$

where $\chi_T(\omega)$ and $\chi_L(\omega)$ are the transverse and longitudinal susceptibilities of the impurity and ω_n is the Larmor frequency. $\Delta K(r)$ is related to $A_s(r)$ by

$$\Delta K(r) = (\hbar^2\gamma_e\gamma_n)^{-1}A_s(r)\chi_{\mathrm{loc}}. \tag{3}$$

The relative average values $\langle A_a\rangle$ on shells B and M are given by the single-crystal data of Stakelon,[11] which showed that $\langle A_a\rangle$ is much smaller on satellite M than on satellite B. As T_1^{-1} values for these satellites scale with $(\Delta K)^2$ rather than $\langle A_a\rangle^2$ at a given T (see Fig. 3), it can be concluded that the scalar process of Eq. (1) dominates the nuclear relaxation. If an isotropic impurity susceptibility

$$\chi_T(\omega) = \chi_{\mathrm{loc}}\frac{1+i\,\omega_e\tau}{1-i(\omega-\omega_e)\tau} \tag{4}$$

is assumed, with a well-defined relaxation time τ (ω_e is the electronic Larmor frequency), Eqs. (1), (3), and (4) yield

$$T_1T(\Delta K)^2 = \chi_{\mathrm{loc}}(2k_B\gamma_n^2\tau)^{-1}(1+\omega_e^2\tau^2). \tag{5}$$

Since in the present temperature range both χ_{loc} and ΔK vary only slightly with T, τ is found from Fig. 3 to be nearly temperature independent. Taking $\chi_{\mathrm{loc}} = 8.30\times10^{-26}$ emu/atom at $T=0$°K,[3] and $T_1T(\Delta K)^2 = 1.90\times10^{-5}$ sec °K^2 for $H=58$ kG yields $\tau = 0.36\times10^{-12}$ sec. The corresponding value $\omega_e^2\tau^2 = 0.15$ is consistent within experimental accuracy with the small decrease of $T_1T(\Delta K)^2$ from 60 to 30 kG. A limiting zero-field value $T_1T(\Delta K)^2 = (1.65\pm0.2)\times10^{-5}$ sec °K^2 can then be deduced.

Such a result can be analyzed by use of the random-phase approximation, which successfully accounts for the impurity NMR data from less-magnetic systems such as *Au*V and *Al*Mn.[1] Within this approximation

$$\tau = \lim_{\omega\to0}\frac{1}{\chi_{\mathrm{loc}}}\mathrm{Im}\left(\frac{\chi_{\mathrm{loc}}}{\omega}\right) = \frac{\pi\hbar\chi_{\mathrm{loc}}}{10\mu_B^2}, \tag{6}$$

which with Eq. (5) ($\omega\tau \ll 1$) yields a Korringa relation $T_1T(\Delta K)^2 = (5\hbar/4\pi k_B)(\gamma_e/\gamma_n)^2 = 5\xi$, where ξ is the Korringa constant. For ^{63}Cu, $5\xi = 1.98\times10^{-5}$ sec °K^2 which is in quite good agreement with the present experimental value for $T_1T(\Delta K)^2$. The small difference might be accounted for by a slight dipolar contribution to the measured T_1^{-1}. This result is basically identical to that which would be obtained for the d contributions to the T_1 and Knight shift of the impurity. It should also be pointed out that, in the absence of a bottleneck, standard ESR techniques would not be able to detect an impurity resonance with such a huge width (about 150 kG from the present value for τ^{-1}).

The present data on the "classic" Kondo system *Cu*Fe support a recent calculation[12] based on a perturbation expansion of the symmetric Anderson Hamiltonian which demonstrated that the Korringa relation [or Eq. (6)] holds for any value of U, for $T \ll T_K$. The same result seems to emerge from numerical calculation of $\tau(T)$ based on the s-d model, which at $T=0$ yields a constant value $\tau^{-1} \sim k_BT_K/\hbar$[13] (the present value for τ would correspond to $T_K \sim 20$°K). Equation (6) indeed implies that both χ_{loc} and τ have the same enhancement factor with respect to the nonmagnetic-impurity case, which means that both are proportional to $(k_BT_K)^{-1}$ for $T \ll T_K$. A further desirable step would be to determine both experimentally and theoretically the temperature dependence of χ_{loc} and τ^{-1}. For χ_{loc}, as for *Au*V, a T^2 deviation from the $T=0$ value has been evidenced here and has been derived by renormalized random-phase-approximation techniques.[14] Although there is no definitive theoretical argument, the present results allow us to conclude safely that any deviation from a linear relation between ΔK and χ_{loc} does not exceed 3% for $0 < T < 100$°K. It might be conjectured that the T dependence of $\Delta K/K$ given in Fig. 2 is also that of χ_{loc}. Finally

NMR measurements of bulk nuclei in CuMn demonstrated that $\tau^{-1} \propto T$ for $T \gg T_K$ as expected from perturbation theories of the s-d model.[7] Here a constant value for τ has been obtained for $T \ll T_K$ in CuFe. Further measurements at higher T might determine how these two regimes are connected to each other.

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Quantum Electrodynamical Effects in Kerr-Newmann Geometries

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Following the classical approach of Sauter, of Heisenberg and Euler and of Schwinger the process of vacuum polarization in the field of a "bare" Kerr-Newman geometry is studied. The value of the critical strength of the electromagnetic fields is given together with an analysis of the feedback of the discharge on the geometry. The relevance of this analysis for current astrophysical observations is mentioned.

Deruelle and Ruffini[1] have recently pointed out how the positive- and negative-root solutions[2] of test particles in the background field of a collapsed object have to be identified with the classical limit of the "positive" and "negative" energy solutions of a relativistic quantum field.[3] From this immediately follows that in the ergosphere[4] or in the effective ergosphere[5] of a black hole the conditions are encountered for the possibility of particle creation through the Klein process[6] as analyzed in the framework of a flat-space theory by Sauter,[7] Heisenberg and Euler,[8] and Schwinger.[9] In this Letter we present the results of the analysis of the vacuum polarization of a Kerr-Newman[10] geometry with particular emphasis on (a) the limitations imposed by vacuum polarization on the strength of the electromagnetic field of a black hole,[11] (b) the efficiency of extracting rotational[12] and Coulomb energy[13] from a black hole by pair creation, and (c) the possibility of having observational consequences of astrophysical interest. It is important to stress that the processes here analyzed differ in three major respects from the one recently analyzed by Hawking.[14] The process of particle creation in the present framework (1) does not require a time-varying background geometry, (2) can occur for masses of current astrophysical interest ($M \gtrsim 10^{33}$