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DIRECT MEASUREMENT OF THE RUDERMAN-KITTEL INTERACTION IN PLATINUM ALLOYS*

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The dipolar interaction between nuclear spins is responsible for the broadening of the nmr absorption lines in solids.[1] In metals Ruderman and Kittel[2] showed that the conduction electrons can give rise to an indirect interaction between nuclear spins of the form $J_{ik}\vec{I}_i\cdot\vec{I}_j$, where $\vec{I}_i$ and I_j are nuclear spin operators. This type coupling also exists between electronic spins and is known to play an important role in magnetism.[3] So far the magnitude of the coupling constant J_{ik} has always been derived indirectly, e.g., by comparing the nmr absorption[4] or dispersion[5] linewidth to a theoretical value based on dipolar coupling alone, or by observing the variation of the relaxation time T_1 in low magnetic fields.[6] Here a method is presented where J can be read directly as the frequency of an oscillatory term in the spin-echo envelope observed in alloys. Various Pt solid solutions have been investigated as an illustration, and experiments are presently under way to determine whether the method can be applied also to other heavy elements.

The nmr absorption signals of ^{195}Pt in solid solutions are broadened[7] because of the existence of a spatial distribution of Knight shifts related to oscillations of the charge density of the conduction electrons.[8] Such a static broadening should not contribute to the decay of the spin-echo envelope.[9] The most striking aspect of the observed spin-echo envelopes, as seen in Fig. 1, is the strong oscillation modulating the decay. Such oscillations were present in all samples studied: solid solutions with, respectively, 6%, 10%, 70% Au in Pt, and 10% Ir in Pt. Their frequency does not depend upon applied magnetic field (range 3 to 10 kG), temperature (1.4 to 4.2°K), or pulse conditions. Within experimental error this frequency was the same in all three low-concentration samples, and somewhat larger for the 70% Au sample. This suggests that it is not determined by the solute nuclei, Au or Ir. We will see, however, that the presence of the solute nuclei is a necessary condition for the existence of the oscillations. The latter can be predicted on the basis of a model analogous to that proposed by Hahn and Maxwell[10] for liquids and their frequency is a direct measure of the Ruderman-Kittel indirect spin-spin interaction.

To show this, let us make the following approximations: (a) We neglect the dipolar and pseudo-dipolar interactions. The strength of the dipolar interaction between nearest neighbors in Pt cannot exceed 130 c/sec, which is small compared with the frequency of the oscillations (~4 kc/sec). (b) We neglect the nonsecular components of the indirect interaction [namely, $\sum_{i<j}J_{ij}(I_i^xI_j^x+I_i^y\times I_j^y)$]. This approximation is justified when the local inhomogeneous broadening is much larger

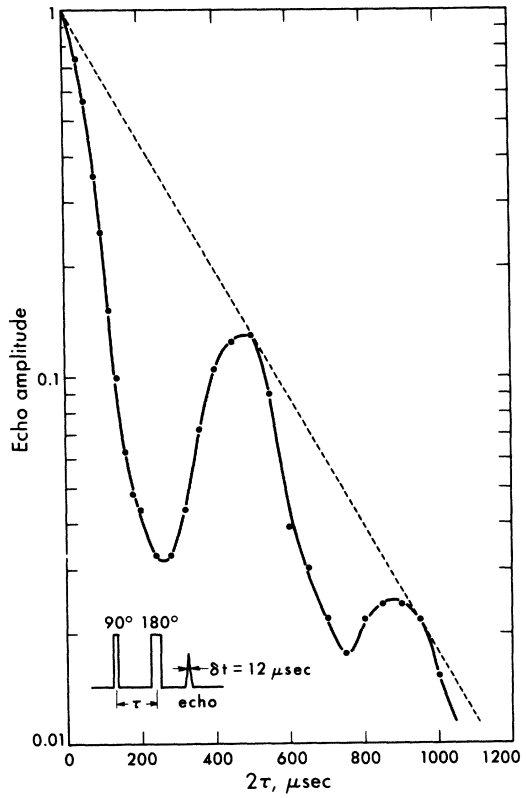

FIG. 1. Variation of the spin-echo amplitude of ^{195}Pt, in a 6 % Au-94 Pt alloy, as a function of the time separating the first rf pulse from the echo. These data were obtained in a homogeneous external field of 10 kG at 1.4°K. The decay time T_2 characterizing the dashed line is 250 μsec, as compared to the 12-μsec width of a single echo. This difference is explained by a local distribution of Knight shifts. The full line was drawn through the experimental points and represents the oscillating spin-echo envelope.

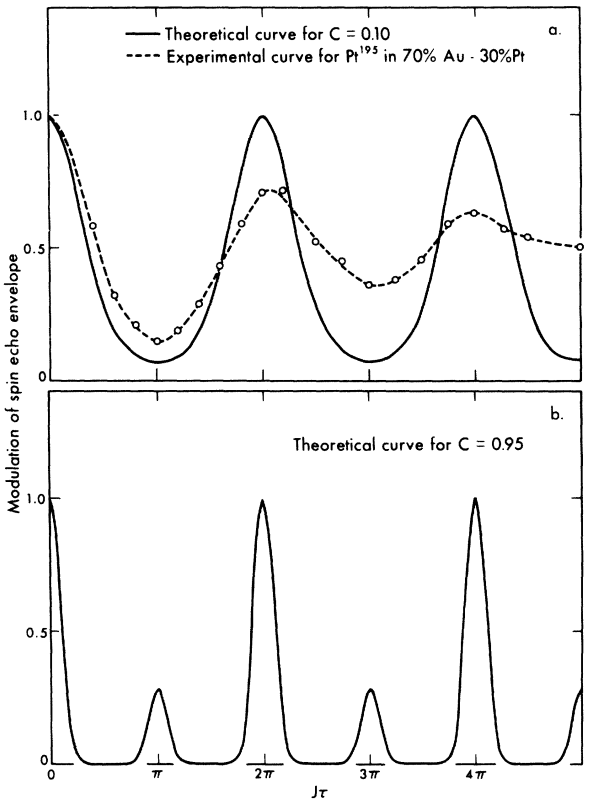

FIG. 2. Oscillating function $E(J\tau)$ modulating the spin-echo envelope. (a) Theoretical and experimental curve for a concentration of the investigated nuclear species (here ^{195}Pt) of 10 %. The damping observed experimentally is not accounted for in our simple model. It may be due to a partial breakdown of assumptions (a), (b), and (c). (b) For high concentrations c, the function exhibits small peaks between the sharp peaks at $J\tau = 2n\pi$. This cannot be checked with ^{195}Pt which has a natural abundance of 33.7 %, but could be verified with isotopically enriched Pt samples, or with a different nuclear species.

than the indirect interaction,[11] a condition that is satisfied here because the alloying produces a local distribution of Knight shifts resulting in an inhomogeneous broadening of order 50 kc/sec. (c) We consider only the indirect interaction between nearest neighbors. This approximation is not essential, but it simplifies the calculation slightly. The amplitude of the spin echo following a $\pi/2$-π pulse sequence, under assumption (b), is proportional to[11]

$$E(2\tau) = \mathrm{Tr}\{\exp(-2i\tau \sum_{i<j} J_{ij} I_i^z I_j^z) \times \sum_k I_k^x \exp(2i\tau \sum_{l<m} J_{lm} I_l^z I_m^z) \sum_n I_n^x\}, \quad (1)$$

where τ is the time interval between both pulses. Because of (a) and (c), $J_{ij} = J$ for nearest neighbors and vanishes otherwise. A straightforward calculation (for $I = 1/2$) yields

$$E(2\tau) = \sum_{r=0}^{N} A_r \cos^r(J\tau), \quad (2)$$

where A_r is the probability of a ^{195}Pt nucleus to have r nearest ^{195}Pt neighbors, and N is the total number of nearest neighbors, 12 for the Pt fcc lattice. For a random distribution, A_r is given by the binomial coefficient

$$A_r = \binom{12}{r} c^r (1-c)^r, \quad (3)$$

where c is the concentration of ^{195}Pt nuclei. When $J\tau$ is an integral multiple of 2π, $E(2\tau)$ attains its maximum value of 1. When $J\tau$ is an odd multiple of π, $E(2\tau)$ attains a low value which is very close to zero unless c is very close to 0 or to 1. Thus we see that the spin-echo amplitude, as function of τ, oscillates at a frequency J without change of its rf phase. Expression (2) has been evaluated by a computer. The result for $c = 0.10$ is shown in Fig. 2 and compared with the experimental data. The use of a phase-coherent spectrometer[12] made it possible to verify that the rf phase of the echo indeed does not change when the amplitude gets very small. Thus we see that our approximation accounts very well for the observed features of the oscillations. When approximation (c) is relaxed, the oscillations at frequency J are found to be modulated by oscillations at frequencies J_2, J_3, etc., where J_2, J_3 are the interaction strengths with next-nearest neighbors, next-next-nearest neighbors, etc. When approximation (a) is relaxed, an expansion in powers of $\mathcal{H}_{\text{dipolar}}/J$ shows that to the first order in this quantity, the shape of the echo envelope is unaffected.

The values of J derived from the experiments are shown in the table:

Sample	Nearest-neighbor coupling constant $J/2\pi$
6% Au-94% Pt	4.14 kc/sec ± 2%
10% Au-90% Pt	4.18 kc/sec ± 2%
70% Au-30% Pt	4.95 kc/sec ± 2%
6% Ir-94% Pt	4.0 kc/sec ± 10%

It will be noted that these values exceed that obtained indirectly in the pure metal[6] by a factor of about 2.

To conclude let us state that the method presented here will be applicable to metals where J is strong compared with the dipolar coupling. Furthermore, J has to be smaller than the difference between the frequencies of neighboring spins, a condition obtained here by alloying. Heavy metals with close-packed structures represent the natural candidates of future investigations. An accurate knowledge of the strength of the Ruderman-Kittel interaction will add to the other experimental data with which one can now try to determine the various parameters entering in the theory of magnetism in metals.

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