from the equilibrium one. We believe this effect is a very useful tool for studying the impurity states and the profile of the hot-electron distribution function under strong magnetic fields. Furthermore, we have information of the cyclotron-resonance absorption in the pulsed electric field.[10] Assuming that the emission and absorption bandwidths are nearly equal, we can estimate the bandwidth of the emission line as $\Delta\lambda/\lambda \sim 10^{-2}$ at $\lambda = 119$ μm. Such a narrow bandwidth offers the possibility of application as a tunable far-infrared source.

We would like to thank Dr. Y. Katayama, Dr. E. Yamada, and Dr. N. Kotera for their fruitful discussions on the research of this field, and also Professor H. Kawamura for his constant encouragement.

[1]K. L. I. Kobayashi and E. Otsuka, in *Proceedings of the Eleventh International Conference on the Physics of Semiconductors, Warsaw, Poland, 1972,* edited by the Polish Academy of Sciences (PWN—Polish Scientific Publishers, Warsaw, 1972), p. 903.

[2]S. J. Bucksbaum, A. G. Chynoweth, and W. L. Feldmann, Appl. Phys. Lett. 6, 67 (1965).

[3]T. Musha, F. Lindvall, and J. Hagglund, Appl. Phys. Lett. 8, 157 (1966).

[4]J. C. Eidson and G. S. Kino, Appl. Phys. Lett. 8, 183 (1966).

[5]G. Bekefi, A. Bers, and S. R. J. Brueck, Phys. Rev. Lett. 19, 24 (1967).

[6]Y. Oka, K. Nagasaka, and S. Narita, Jap. J. Appl. Phys. 7, 611 (1968).

[7]E. Yamada and T. Kurosawa, in *Proceedings of the Ninth International Conference on the Physics of Semiconductors, Moscow, U. S. S. R., 1968* (Nauka, Leningrad, 1968), p. 800.

[8]R. F. Kazarinov and V. G. Skobov, Zh. Eksp. Teor. Fiz. 42, 1047 (1962) [Sov. Phys. JETP 15, 726 (1962)].

[9]N. Kotera, E. Yamada, and K. F. Komatsubara, J. Phys. Chem. Solids 33, 1311 (1972).

[10]K. L. I. Kobayashi and E. Otsuka, Solid State Commun. 11, 85 (1972).

[11]Y. Yafet, R. W. Keyes, and E. N. Adams, J. Phys. Chem. Solids 1, 143 (1956).

Measurement of Spin-Scattering Anisotropy and Exchange-Coupling Energy in Cu-Fe, Using the Wave Shape of the de Haas–van Alphen Effect*

H. Alles,† R. J. Higgins,‡ and D. H. Lowndes
Physics Department, University of Oregon, Eugene, Oregon 97403
(Received 29 January 1973)

A linearly field-dependent and temperature-independent exchange energy and spin-scattering anisotropy has been measured in a Cu–93-ppm Fe alloy using wave-shape analysis of the de Haas–van Alphen effect for the ⟨111⟩ neck orbit over a field range of 30–45 kG and a temperature range of 1–2°K.

Landau quantum oscillations such as the de Haas–van Alphen (dHvA) effect provide an explicit measure of the interaction between conduction electrons and magnetic impurities. In contrast to bulk-property measurements[1] (resistance, NMR, etc.), only specific orbits are observed and comparison[2] with theory is more direct. Analysis of the amplitudes gives the local scattering rate directly,[3] and analysis of the wave shape gives spin-dependent information. Recently, Coleridge, Scott, and Templeton[4,5] have observed a shift in angular position of the dHvA spin-split zero in Cu doped with Cr and other transition-metal impurities, and have interpreted their data to obtain the sign (negative or antiferromagnetic) and estimated magnitude of the impurity-electron exchange energy $\epsilon_{ex} = \mu_B H_{ex}$. They also observed in some cases the disappearance of the spin-split zero, which they interpreted as due to anisotropy of scattering of spin-up and spin-down electrons by the magnetic impurity.[5]

We have developed a more general technique[6] using dHvA wave-shape analysis for the measurement of spin scattering anisotropy (SSA) and exchange energy, which does not require the (accidental) spin zero. The dHvA signal is resolved into the harmonic components contributed by spin-up and spin-down electrons. Since the harmonic amplitudes from each spin may be unequal in the presence of a magnetic impurity, the resultant wave shape is altered in magnitude and phase from the usual Lifshitz-Kosevich (LK) expression.[7]

Consider the four most dominant contributions to the dHvA magnetization M_r^{σ}, where σ is the spin index and r is the harmonic index. Using

the Lifshitz-Kosevich theory,[8] $M_r{}^{\circ}$ may be expressed as

$$M_r{}^{\circ}=\frac{\sqrt{H}Dx_r}{r^{3/2}\sinh(x_r)}\exp(-r\alpha X^{\circ}/H)\times\sin[2\pi r(F/H-\gamma)\mp\pi/4],$$

where $x_r=r\alpha T/H$, X° is the scattering temperature, D is a geometrical factor, F is the frequency, γ is the infinite field phase, and $\alpha=146.9\times m^*/m$ kG/°K. If we move quantities not involving X° to the left-hand side of the equation, a more convenient amplitude $\tilde{M}_r{}^{\circ}$ may be defined such that

$$\tilde{M}_r{}^{\circ}=\begin{cases}B, & \text{for the first dHvA harmonic, spin up;}\\ BA, & \text{for the first dHvA harmonic, spin down;}\\ B^2, & \text{for the second dHvA harmonic, spin up;}\\ B^2A^2, & \text{for the second dHvA harmonic, spin down;}\end{cases}$$

$$B=\exp(-\alpha X^0/H),$$

$$A=\exp(-\alpha\delta_X/H),$$

where $X^+=X^0$ and $X^-=X^0+\delta_X$. The up and down contributions will be out of phase with each other by an amount

$$\Phi=\pi g'm^*/m,$$

where $g'=g-\epsilon_{ex}/\mu_B H=g-H_{ex}/H$.

The components can be conveniently combined into first- and second-harmonic resultants R_r using the phasor diagrams in Fig. 1. Since the up and down components are unequal, the resultant suffers a phase shift θ_r. Elementary trigonometric manipulations yield

$$R_1=B(1+A^2+2A\cos\Phi)^{1/2},$$

$$R_2=B^2(1+A^4+2A^2\cos2\Phi)^{1/2},$$

$$\theta_1=\tan^{-1}[\tan(\Phi/2)(1-A)/(1+A)],$$

$$\theta_2=\tan^{-1}[\tan(\Phi)(1-A^2)/(1+A^2)].$$

If φ_r is the phase of the rth dHvA harmonic relative to an arbitrary field value H_0 (found by using Fourier analysis on high-resolution data), then $2\varphi_1-\varphi_2$ will be independent of H_0 and equal to $2\theta_1-\theta_2\pm\pi/4$. Two other quantities may also be computed from the data, $\ln(R_2/R_1)$ and $d(\ln(R_2/R_1))/d(1/H)$, where ratios are used to eliminate a variety of experimental complications. Using

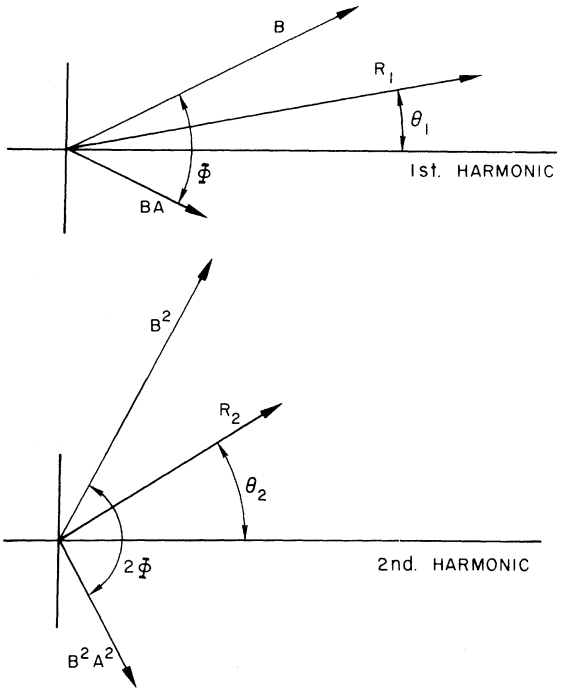

FIG. 1. Phasor diagram of the first and second dHvA harmonic amplitudes which result from the contribution from spin-up and spin-down electrons. Φ is the spin-splitting angle and the phase shifts θ_1 and θ_2 are caused by the spin-scattering anisotropy.

these computed quantities, a parameter fit to the data yields the quantities of interest, Φ, X^0, and δ_X, at any given value of magnetic field and temperature.

In Fig. 2 are shown the results of measurements on the neck orbit ($H\|[111]$) in a Cu crystal with 93 ppm Fe (measured by atomic absorption). The measurements were made using a computer-assisted system[7] with which it was possible to control or eliminate a variety of potential errors in the measurement of dHvA harmonic amplitudes and phases. Of particular importance was simultaneous phase-sensitive detection at multiple (up to 7) harmonics and quadrature phases of the modulation frequency, which allowed accurate measurement of weak (1 part in 2000) dHvA harmonic content and the elimination of wave-shape errors due to eddy currents.

Relative phase measurements in a pure Cu crystal gave a value of $2\varphi_1-\varphi_2$ of $-45\pm1°$, in agreement with the Lifshitz-Kosevich theory.[8] This value would have been altered if magnetic interaction (MI) effects were important.[9,10] Using the pure Cu relative phase for the neck orbit as a reference, the net impurity-induced phase *shift*

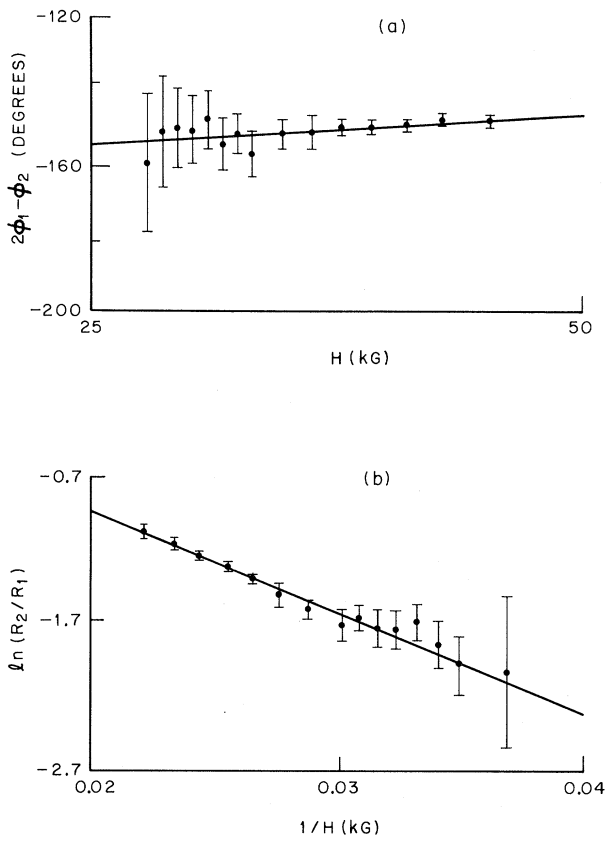

FIG. 2. (a) Relative phase $2\varphi_1-\varphi_2$ as a function of the applied field in Cu–93-ppm Fe at 1.23°K. The measured value in pure Cu was $-45°$, the Fe impurity introducing a shift of approximately $-105°$. Although $2\varphi_1-\varphi_2$ is determined modπ, a comparison between Cu and Cu-Fe data taken under identical conditions allows the shift $2\theta_1-\theta_2$ to be evaluated mod2π. The larger error bars at low field are a result of a rapidly decreasing record harmonic amplitude. (b) $\ln(R_2/R_1)$ as a function of $1/H$ at 1.23°K. The value of R_2/R_1 is found from the measured harmonic amplitude ratio as described in the text.

(Fig. 2) is about $-105°$. In the limit of total SSA, the value would be 180°. From the sign of the shift, it can be deduced that spin-up electrons are scattered more strongly than spin-down electrons.[11] A summary of results of measurements at a temperature of 1.2°K and of computed theoretical parameters defined above is given in Table I. Our results are in general agreement with those of Coleridge, Scott, and Templeton[5] using the spin-zero technique. However, comparisons involve additional theoretical and experimental complications beyond the scope of this Letter.

Measured and calculated quantities are listed in Table I. The values of H_{ex} and δ_X scale linearly with field, and extrapolate to zero in the $H=0$ limit. It is of interest to compare these results with several recent calculations.[12-14]

We note that for Cu-Fe, we are in the low-temperature, low-field regime of the Kondo ground state, since the experimental range of field and temperature covers

$$2.5<[(k_BT)^2+(\mu_BH)^2]^{1/2}<4.6<(k_BT_K\geq 10\ \text{K}).$$

Simpson and Paton[12] have developed expressions for the quantities δ_X and Φ measured in this experiment:

$$k_B\delta_X=2cJ^2\rho\langle S_z\rangle(1+\text{a log term}),$$

$$\Phi_{Cu}-\Phi_{Cu\text{-}Fe}=\frac{\pi m^*}{m}\frac{cJ\langle S_z\rangle}{\mu_BH}(1+\text{a log term}).$$

Although these are not expected to be precisely correct in the $T<T_K$ regime since the calculation was only to third order in perturbation theory, a more exact calculation is likely to alter only the log term. We note that the data agree with the theory in that the dominant H dependence is contained in the $\langle S_z\rangle$ term. Mössbauer measure-

TABLE I. Measured quantities and derived quantities discussed in the text for 93-ppm Cu-Fe at 1.23°K. The logarithmic derivative (column 3) is similar to a conventional scattering temperature, and is defined as $146.9(m^*/m)\times d(\ln(R_2/R_1))/d(1/H)$. We observe no appreciable temperature dependence (within the approximate uncertainties quoted in the table, bottom line) in $2\theta_1-\theta_2$ and the calculated quantities over a temperature range from 1.2 to 2.1°K.

Field (kG)	$2\theta_1-\theta_2$ (deg)	$\ln(R_2/R_1)$	Logarithmic derivative (°K)	$\Phi_{Cu}-\Phi_{Cu\text{-}Fe}$ (deg)	H_{ex} (kG)	X^0 (spin down) (°K)	δ_X (°K)	$X^0+\delta_X$ (spin up) (°K)
30	−107.6	−1.86	1.02	12.6	4.55	1.09	0.28	1.37
45	−102.0	−1.09		12.6	6.8	1.14	0.38	1.52
	±2	±0.1	±0.1	±1.5	±0.8	±0.08	±0.03	±0.1

ments[15] in Cu-Fe indicate that the polarization $\langle S_z \rangle$ of the Fe moment is linear in H to fields well above 50 kG, in agreement with our observation that both H_{ex} and δ_X are linear in H. We note that the scattering rate for spin-down electrons is nearly field independent, while the rate for spin-up electrons increases with applied field. This is opposite to the small negative bulk magnetoresistance. Further interpretation of this difference in terms of the details of the Cu Fermi surface, details of the impurity level, and differences between the scattering rates measured by dHvA and by resistivity, are beyond the scope of this note.

The wave-shape analysis technique outlined here may be used for other orbits, and is particularly useful in systems without convenient spin zeros (Au-Fe, for example). The ability to resolve individual spins and follow the individual field and temperature dependence provides additional information on the impurity interaction not available by other techniques.

We thank F. Haberda, Y. K. Chang, K. Bachman, and D. Malm for assistance with various phases of crystal preparation and chemical analysis (the latter carried out at Bell Laboratories); J. Nosler for helpful discussions; and especially P. Coleridge and I. M. Templeton for a stimulating discussion prior to publication of their exchange-splitting data, which led to the choice of Cu-Fe as the first test for the technique described here.

*Research conducted at the University of Oregon with the support of the National Science Foundation.

†Current address: Bell Laboratories, Murray Hill, N. J. 07974.

‡Address through August 1973: Bell Laboratories, Murray Hill, N. J. 07974.

[1]For a review, see A. J. Heeger, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1969), Vol. 23.

[2]M. T. Beal-Monod and R. A. Weiner, Phys. Rev. 170, 552 (1968).

[3]M. Springford, Advan. Phys. 20, 493 (1971); D. W. Terwilliger and R. J. Higgins, J. Appl. Phys. 43, 3346 (1972).

[4]P. T. Coleridge and I. M. Templeton, Phys. Rev. Lett. 24, 108 (1970).

[5]P. T. Coleridge, G. B. Scott, and I. M. Templeton, Can. J. Phys. 50, 1999 (1972).

[6]Details may be found in H. Alles, thesis, University of Oregon, 1972 (unpublished), and to be published.

[7]The notion of unequal dHvA amplitudes for electrons with opposite spins and the consequent dHvA phase shift was advanced earlier in discussing a temperature-dependent phase in pure Zn. See W. J. O'Sullivan and J. E. Schirber, Phys. Rev. 162, 519 (1967).

[8]I. M. Lifshitz and A. M. Kosevich, Zh. Eksp. Teor. Fiz. 29, 730 (1955) [Sov. Phys. JETP 2, 636 (1956)]. For a discussion including scattering, see A. V. Gold, in *Solid State Physics: Electrons in Metals*, edited by J. F. Cochran and R. R. Haering (Gordon and Breach, New York, 1968), Vol. I.

[9]D. Shoenberg *et al.*, in *Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, Japan, 1970,* edited by E. Kanda (Keigaku Publishing Co., Tokyo, 1971), p. 51.

[10]R. A. Phillips and A. V. Gold, Phys. Rev. 128, 932 (1969).

[11]We follow the convention that electron spin and magnetic moment point oppositely, with the lowest energy state in a magnetic field being magnetic moment up (+) and spin down (−). See, for example, C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1966), 3rd ed., p. 435.

[12]G. Simpson and B. E. Paton, Phys. Lett. 34A, 180 (1971).

[13]H. Miwa, T. Ando, and H. Shiba, Solid State Commun 8, 2161 (1970).

[14]E. W. Fenton, Solid State Commun. 10, 185 (1972).

[15]R. B. Frankel, N. A. Blum, B. B. Schwartz, and D. J. Kim, Phys. Rev. Lett. 12, 105 (1967); T. A. Kitchens, W. A. Steyert, and R. D. Taylor, Phys. Rev. 138, A467 (1965).