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Temperature Dependence of Hyperfine Fields in Dysprosium Intermetallic Compounds; $DyCo_5$, $DyNi_5$

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The Mössbauer technique is used to investigate the temperature dependence of the effective fields acting on Dy^{161} nuclei in two intermetallic, Cu_5Ca-structure-type compounds. From these measurements the temperature dependence of the dysprosium sublattice magnetization is derived. The results are compared with macroscopic magnetization measurements and with the predictions of theoretical models. In the case of $DyCo_5$ the molecular-field model gives a very good approximation. The exchange field acting on Dy spins at 0°K is estimated to be $\beta_0 H_{\mathrm{exc}}/K = 142°\mathrm{K}$. The value of the magnetic hyperfine interaction at 4°K in $DyCo_5$ is $g_0\mu_n H_{\mathrm{eff}} = -862 \pm 15$ Mc/sec and in $DyNi_5$ it is -790 ± 25 Mc/sec. The value of the effective field acting on Dy^{161} nuclei in $DyNi_5$ corresponds to an ionic magnetic moment of $9.3 \pm 0.3\mu_B$. This value is higher than that obtained by magnetic measurements ($8.6\mu_B$) and might indicate that the Ni sublattice contributes to the total magnetization by an antiferromagnetic coupling.

INTRODUCTION

In rare-earth compounds the effective magnetic field (H_{eff}) acting on the rare-earth nuclei is generally proportional to the ionic magnetic moment (M_s) and thus the temperature dependence of H_{eff} follows that of M_s.[1] This proportionality may be destroyed by several effects:

(a) Mixing of ionic excited states into the ionic ground state. This effect is very large in the case of Eu[2] and Sm[3] where the first excited states are relatively low and completely destroy the proportionality between H_{eff} and M_s. In the heavy rare-earth ions this effect would disturb the proportionality between H_{eff} and M_s to less than 2%.

(b) The contribution to H_{eff} arising from core polarization. This is large relative to the main $4f$ contributions in the cases of Sm, Eu, and Gd. In the heavy rare-earth elements this contribution is less than 5% and to first approximation its value is proportional to $\langle J_z \rangle$,[4] thus following the temperature dependence of M_s.

(c) The contribution to H_{eff} arising from polarized conduction electrons will not follow the temperature dependence of M_s, especially if the polarization is mainly caused by other than the rare-earth magnetic ions themselves, as in rare-earth intermetallic compounds containing iron or cobalt. This contribution to H_{eff} appears to be large in rare-earth intermetallics with iron.[5]

In dysprosium intermetallic compounds (excluding those containing iron) all the above-mentioned effects will have small influence (less than 3%) on the proportionality between H_{eff} and M_s. Thus the Mössbauer technique, by which H_{eff} can be measured to high accuracy, is a powerful tool in measuring the rare-earth sublattice magnetization and its temperature dependence. For this purpose, there are several advantages in using the Mössbauer effect over conventional bulk-magnetization measurements:

(1) By the Mössbauer effect one can measure the magnetization of a single site or sublattice, whereas the bulk-magnetization measurement gives information only on the total sum of the magnetic moments of all the sites in the crystal.

(2) By the Mössbauer effect one can investigate the properties of several inequivalent magnetic sites of the same ion, as was done for thulium iron garnet.[6] Magnetic measurements cannot identify inequivalent magnetic sites.

(3) The values of the magnetic moments measured by bulk magnetization measurements are generally lower than their actual values because of anisotropy effects especially if the experiments are done with polycrystalline samples. The magnetic fields in which such experiments have been done, were in many cases too low to reach saturation values near 0°K. Such a difficulty does not exist in a Mössbauer measurement which is a microscopic measurement. Macroscopic anisotropy effects have no influence on the results.

(4) The applied magnetic field in a bulk magnetization measurement sometimes destroys or changes the magnetic structure of the compound and thus the results obtained are completely different from the values at zero field.

(5) In an antiferromagnetic compound bulk magnetization measurements give no information about the moments and their relative orientation on the two sites coupled antiferromagnetically. Mössbauer experiments, because of their microscopic nature, measure the magnetic moment on a single ion, thus enabling one to measure the magnetic moments on the various sites in antiferromagnetic compounds.

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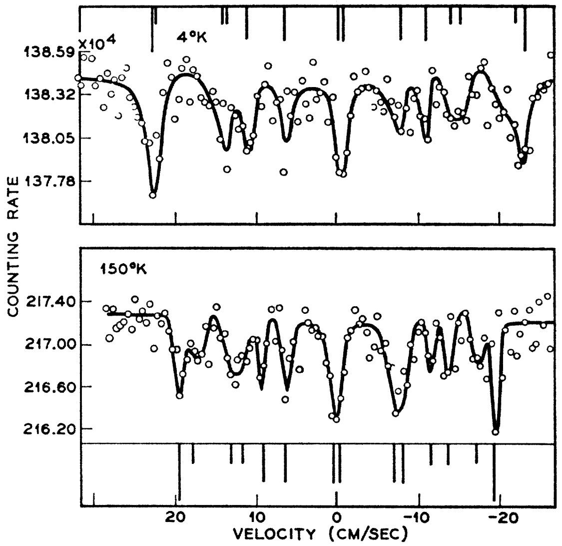

FIG. 1. Mössbauer absorption spectra of $DyCo_5$. The calculated positions and theoretical intensities are given below.

(6) Bulk magnetization measurements are made in a magnetic field and the zero-field value is found by extrapolation, whereas the Mössbauer technique measures the zero-field value directly.

(7) In bulk magnetization measurements small quantities of magnetic impurities or different phases and small deviations from stoichiometry in the sample investigated may cause large changes in the measured magnetic moments. Again, because the Mössbauer measurement is a microscopic measurement it is much less sensitive to impurities.

The considerations given above indicate that the Mössbauer technique may make major contributions to investigations of magnetic properties of rare-earth ferrimagnetic compounds.

In this paper we report measurements of the temperature dependence of the hyperfine fields (H_{eff} and the electric field gradient q_{eff}) acting on nuclei of Dy^{161} in the two isostructural compounds $DyCo_5$ and $DyNi_5$.[7] The results are compared with the bulk magnetization measurements[8] and with theoretical models. It turns out that the molecular-field model is a very good approximation in the case of $DyCo_5$. The exchange field acting on dysprosium spins at 0°K was estimated to be $\beta_0 H_{\mathrm{exc}}/K = 142\pm2$°K. The saturation value of H_{eff} in $DyCo_5$ is somewhat higher (4%) than that in paramagnetic dysprosium consistent with results in $DyFe_2$,[5] $DyCo_2$,[9] and $DyFe_4$.[10] The value of H_{eff} (4°K) in $DyNi_5$ is 95% of that in Dy metal, and it corresponds to a magnetic moment of $9.3\,\mu_B$. This result when compared to the bulk magnetization results indicates that probably the nickel sites contribute to the total magnetic moment of $DyNi_5$ by an antiferromagnetic coupling.

EXPERIMENTAL DETAILS AND RESULTS

The Mössbauer technique was used to investigate the hyperfine interactions in the ground- and 25.7-keV state of naturally abundant Dy^{161} in $DyCo_5$ and $DyNi_5$. The experimental setup is similar to that described by Cohen *et al.*[11] The source was in the form of Gd_2O_3 at room temperature. The absorbers ($\sim$60 mg/cm^2) were put in a commercial cryostat. A heating coil was used to reach the desired temperatures, which were measured by a thermocouple. Measurements were performed at 4, 58, 98, 147, 192, 250, and 300°K in the case of $DyCo_5$, and at 4, 20, 77, and 300°K in the case of $DyNi_5$. Some of the experimental spectra are given in Figs. 1 and 2.

The analysis of the experimental spectra was greatly simplified by the fact that the ratios of the magnetic and quadrupole moments of the first excited and ground state of Dy^{161} are accurately known ($g_{\mathrm{exc}}/g_0 = -1.21 \pm 0.02$ $Q_{\mathrm{exc}}/Q_0 = 0.98\pm0.03$).[12] Each nuclear level is split by the effective fields according to

$$E=\Delta E-gI_z+\epsilon\left(\tfrac{3}{2}I_z^2-\tfrac{1}{2}I(I+1)\right).$$

Using the ratios $g_{\mathrm{exc}}/g_0=-1.2$, and $Q_{\mathrm{exc}}/Q_0\approx1$, the transition energies were plotted as a function of ϵ/g_0 (Fig. 3). The best resolved spectrum is expected at

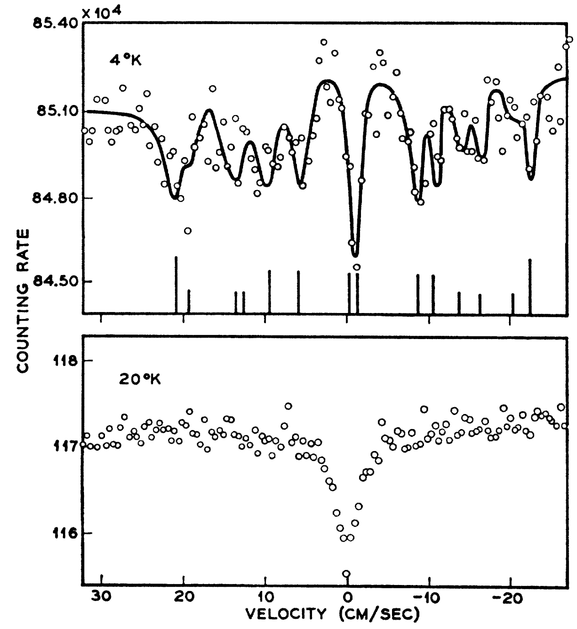

FIG. 2. Mössbauer absorption spectra of $DyNi_5$. The calculated positions and theoretical intensities are given below.

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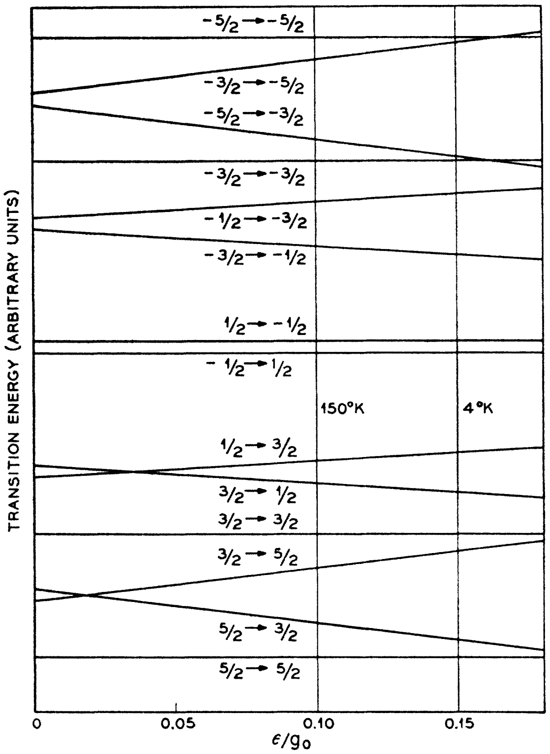

FIG. 3. The transition energies in Mössbauer spectra of Dy^{161} as a function of the quadrupole divided by the magnetic hyperfine constants.

$\epsilon/g_0 \approx 0.1$. The hyperfine splitting of the two nuclear states and the absorption transitions for $\epsilon/g_0 \approx 0.1$ are given in Fig. 4. The ratio of 0.1 for ϵ/g_0 is obtained in the case of $\mathrm{DyCo_5}$ at 150°K. On the other hand, at 4°K many absorption lines overlap (see Figs. 1 and 3). The values of $(g_{\mathrm{exc}}+g_0)$ and $(\epsilon_{\mathrm{exc}}+\epsilon_0)$, which are independent of the ratios g_{exo}/g_0 and $\epsilon_{\mathrm{exo}}/\epsilon_0$, were calculated for each spectrum by a least-squares analysis. The values, normalized to the value at 4°K, are shown in Fig. 5. The results obtained with $\mathrm{DyNi_5}$ are shown in Fig. 2. At 77 and 300°K the spectrum is similar to that at 20°K. The saturation values of the hyperfine interactions in $\mathrm{DyCo_5}$ and $\mathrm{DyNi_5}$ are given in Table I. Values obtained for some other compounds are given for comparison.

TABLE I. Hyperfine interaction energies of several dysprosium compounds.

Compound	$\frac{1}{4}eQ_0q_{\mathrm{eff}}(4°\mathrm{K})/h$ Mc/sec	$g_0\mu_nH_{\mathrm{eff}}(4°\mathrm{K})/h$ Mc/sec	H_{eff} relative to the free-ion value[a]
DyAl garnet[a]	370±15	$-765\pm15(g_z=18.2)$ $-830\pm15(g_z=19.7)$	1.00
Dy metal[b]	535±40	−825±25	1.00
$\mathrm{DyAl_2}$[b]	580±80	−830±25	1.00
$\mathrm{DyFe_2}$[b]	675±70	−950±25	1.13
$\mathrm{DyFe_4}$[c]	580±40	−905±15	1.09
$\mathrm{DyCo_5}$[d]	630±40	−862±15	1.04
$\mathrm{DyNi_5}$[d]	420±80	−790±25	0.95

[a] Reference 12.
[b] Reference 5.
[c] Reference 10.
[d] Present measurement.

DISCUSSION

In $\mathrm{DyCo_5}$, the Dy-Dy exchange interactions are small in comparison to the Dy-Co exchange interactions. This appears to be true because in the isostructural $\mathrm{DyNi_5}$, where the Ni contributes very little to the magnetic ordering, the Néel point is very low (15°K). Thus it is tempting to fit the experimental results of $H_{\mathrm{eff}}(T)$ to the molecular-field-model approximation in which only an exchange field proportional to the cobalt sublattice magnetization acts on the dysprosium spins. Thus the upper solid curve in Fig. 5 is a Brillouin function where $\beta_0H_{\mathrm{exo}}(0)/K = 142°\mathrm{K}$ and H_{exo} follows the temperature dependence of the cobalt sublattice magnetization. The agreement with the experimental points is excellent which indicates the validity of the use of the molecular-field model for this and for similar compounds.[13]

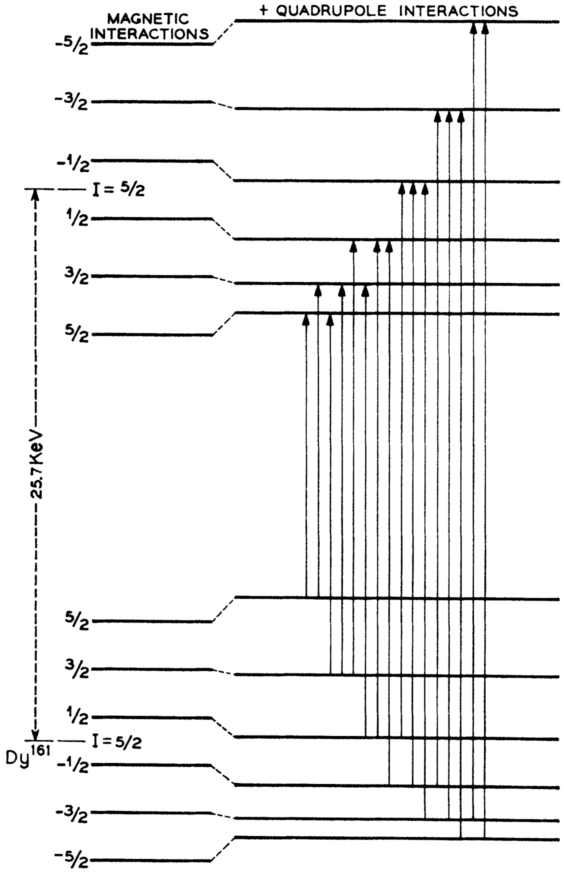

FIG. 4. The hyperfine splitting of the ground and first excited state of Dy^{161}. The allowed nuclear transitions are indicated.

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[13] R. L. Cohen, Phys. Rev. 134, A94 (1964).

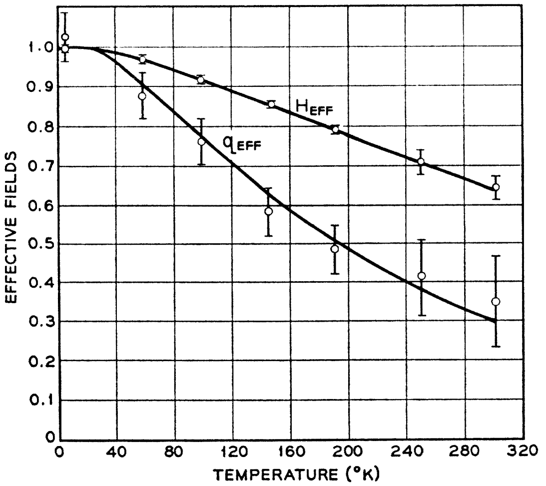

FIG. 5. Theoretical and experimentally measured temperature dependence of the effective fields acting on Dy^{161} in DyCo$_5$.

In the rare-earth ferromagnetic compounds, the electric-field gradient acting on the rare-earth nuclei comes mainly from the $4f$ orbitals oriented by the exchange interactions. Thus the temperature dependence of the quadrupole interactions must follow a modified Brillouin function—a thermal average of $\langle 3J_z^2 - J(J+1)\rangle$.[14] The lower solid line in Fig. 5 is the theoretical temperature dependence of the quadrupole interaction using the value of H_{exc} derived from the temperature dependence of H_{eff}. The agreement with the experimental points is good, which shows that q_{eff} arises mainly from the $4f$ orbitals.

The saturation value of the magnetic hyperfine interaction in DyCo$_5$ is higher by 4% than that obtained in Dy metal, DyAl$_2$[5] and that obtained in paramagnetic dysprosium compounds.[15] This trend is consistent with the idea that in rare-earth intermetallic compounds with $3d$ transition metals having high magnetic moments, there is an additional contribution to H_{eff}. Similar effects were observed in DyFe$_2$, DyCo$_2$[9] in TmFe$_2$[13] and ErFe$_2$.[16] The origin of the additional field is probably due to the polarization of the conduction electrons by the $3d$ electrons.

It is interesting to compare our results of the temperature dependence of the dysprosium sublattice magnetization in DyCo$_5$ to that obtained by bulk magnetization measurements.[8] The measurements performed on YCo$_5$ and DyCo$_5$ indicate that in the range of temperatures between 0 and 300°K the cobalt sublattice magnetization is almost constant ($\sim$8.1 μ_B) and the dysprosium sublattice is coupled antiferromagnetically to the cobalt sublattice. Thus when the magnetic moment of Dy decreases from its saturation value (9.8 μ_B, since $g_J = 1.31$[17]) at a certain temperature ($\sim$150°K) a compensation point is obtained. At this temperature the cobalt sublattice has a moment equal in size to the resultant moment of the Dy sites. According to our Mössbauer measurements, the magnetic moment of the Dy site at 150°K is 8.3 μ_B. This is close to the value of the cobalt sublattice at 150°K in YCo$_5$ (8.0 μ_B). This indicates that the cobalt sublattice in DyCo$_5$ has almost the same moment as in YCo$_5$. The presence of the Dy does not change this moment by more than several percent. If the simple antiferromagnetic coupling between the Dy and Co sites is valid, one should be able to construct the temperature dependence of the bulk magnetization in DyCo$_5$ by simply subtracting from the present measured dysprosium sublattice magnetization, the bulk magnetization of YCo$_5$. Such a procedure leads to the results shown in Fig. 6. The agreement is poor and this may be due to several reasons, some of which were given in the Introduction. They lie in the basic differences between two methods of measurements, i.e., bulk magnetization and Mössbauer effect. However, more importantly, it appears that the disagreement indicates

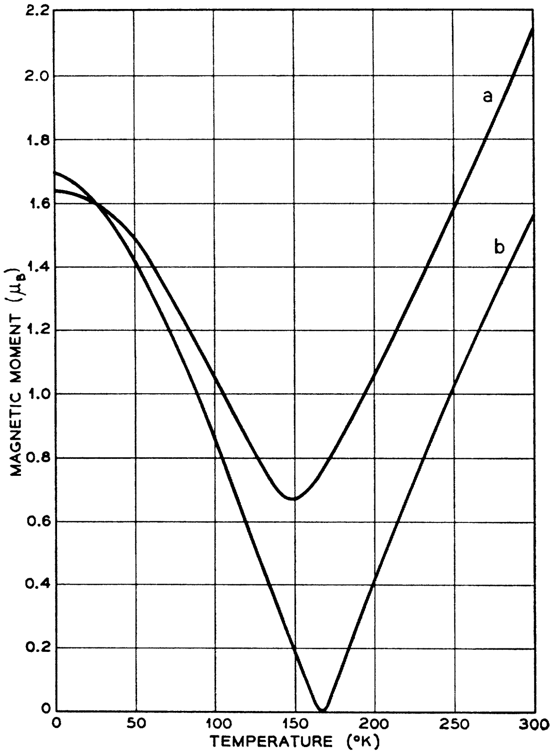

FIG. 6. (a) The experimental temperature dependence of the magnetic moment of DyCo$_5$ (see Ref. 8). (b) The magnetic moment of DyCo$_5$ calculated from the present Mössbauer results assuming a simple antiferromagnetic coupling.

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[15] Recent Mössbauer measurements in DyAlG and DyES (see Ref. 12) have enabled one to estimate $H_{\text{eff}}(0)$ for the state $J_z = J$ which is believed to be the free-ion value.

[16] R. L. Cohen and J. H. Wernick, Phys. Rev. **134**, B503 (1964); H. Dobler, G. Petrich, S. Hüfner, P. Kienle, W. Wiedemann, and H. Eicher, Phys. Letters **10**, 319 (1964).

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in a rather convincing way that the pure antiferromagnetic antiparallel coupling between the Dy and Co sites is an oversimplification. The results indicate that probably the dysprosium magnetic moment is decreasing and changing angle relative to the cobalt magnetic moment when the temperature is raised. Such a picture is in agreement with observed spin structure in $HoCo_5$.[18,19]

The observed effective magnetic field acting on Dy nuclei in $DyNi_5$ is 95% of the free-ion value calculated from paramagnetic compounds. This corresponds to a magnetic moment of 9.3 μ_B. The reported[8] magnetic measurements up to 14 000 Oe yield a value of 7.7 μ_B per formula unit of $DyNi_5$. Because of the lack of saturation, measurements were made in fields up to 80 kOe. A value of 8.6 μ_B was thus obtained.[20] Since at 4°K we might still not be at saturation (the Néel point is 15°K), the value of 9.3 μ_B is a lower limit for the saturated dysprosium magnetic moment. If during the bulk magnetization measurements, saturation was reached, the present results show that the nickel sites in $DyNi_5$ carry a moment above 0.7 μ_B, in contrast to the assumption that the Ni is neutral in this compound, as suggested by the early bulk magnetization measurements[8] on which Bleaney's theoretical investigation[21] was based. The high magnetic moments of Dy^{3+} in $DyNi_5$ and $DyNi_2$[9] suggest that crystalline-field effects may not be important in these crystals.[22]

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[18] Williams James, Remy Lemaire, and Felix Bertaut, Compt. Rend. **255**, 896 (1962).

[19] In $HoCo_5$, neutron-diffraction measurements (see Ref. 18) show that the Co moments lie along the C axis at 4.2 and 300°K, while the Ho moments are at 22° to the C axis at 4.2°K and 90° to the C axis at 300°K.

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Linear-Network Model for Magnetic Breakdown in Two Dimensions*

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A heuristic model is set up for magnetic breakdown in a two-dimensional rectangular lattice at right angles to an applied magnetic field. The model is an adaptation of a method due to Pippard to a set of coupled ordinary differential equations derived from the Schrödinger equation. A linear-chain network is set up. This network can also be derived from Pippard's two-dimensional network and it suggests a simple way to compute the energy bands not only when the number F of flux quanta through a unit cell is the reciprocal of an even integer (the case treated by Pippard) but also when F is any rational fraction. Numerical computations of the energy bands suggest that in the latter case the electron wave moves on large orbits which might be called "hyperorbits." These hyperorbits may be open in a rectangular lattice and may give a resonant open-orbit ultrasonic attenuation. It is also found that when a free-electron Landau level is broadened by the lattice, it splits into two bands separated by a gap. This gap moves through the states from the Landau level as F is changed and may give rise to new de Haas–van Alphen periods. The physical cause of this gap is discussed.

1. INTRODUCTION

RECENTLY, Pippard[1,2] has considered the problem of magnetic breakdown[3] in terms of the Bragg reflection of an electron moving in circular orbits in a magnetic field. An electron may be flipped from one orbit to another and its motion is confined to a periodic network of coupled orbits. Phase memory is not lost in these reflections and the phase coherence must be taken into account. In between the points where Bragg reflection can take place the effect of the lattice is ignored and the wave functions are taken to be those of a free electron in a magnetic field. The strength of the reflections is a parameter in the calculation. If this parameter is zero the model represents a situation of total breakdown. If it is unity the model represents approximately the quasiclassical situation without breakdown.

Pippard's models describe the motion of an electron in a two-dimensional "metal" with a uniform magnetic field perpendicular to its plane. The states are labelled

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