Low-temperature properties of amorphous magnetic materials with random axis of anisotropy

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In amorphous magnetic materials containing heavy rare-earth ions, the single-ion anisotropy energy is as a rule of the order of or larger than the exchange energy. It is shown that because of the large spin of the rare-earth ions, the splitting of the doubly degenerate (in the absence of a molecular field) ground state is small for spins whose local axis of anisotropy is almost perpendicular to the molecular field. An assembly of such two-level systems, with fluctuating splitting energy and transition probability, is similar to the model proposed by Anderson, Halperin, and Varma to explain the anomalous properties of glasses. Transitions between weakly split levels lead to a linear temperature dependence of specific heat and of the electrical resistivity and to strong absorption of ultrasound. The absorption coefficient of ultrasound is frequency-independent over a wide frequency range.

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1. INTRODUCTION

Amorphous intermetallic compounds (Metglas type alloys), one of whose components is a rare-earth metal (Dy, Tb),[1] are magnetic materials with a random axis of anisotropy (RAA). The uniqueness of the magnetic properties of these materials is primarily determined by the fact that for the spins of the rare-earth ions, the direction of the local axis of anisotropy varies randomly in space, and that the anisotropy energy is comparable with or larger than the exchange energy.[1-3] The latter fact leads to the result that the spins are directed principally along the local axes of anisotropy. Below the Curie temperature, each spin is directed at an acute angle to the local molecular field. The resulting magnetic moment is smaller than the moment in a crystalline magnet of the same composition, since even at low temperatures the spins are not parallel to each other. For the same reason, the Curie temperature of amorphous compounds is found to be considerably lower than of crystalline. Because of the large anisotropy energy and of the random orientation of the axes, the coercive force is anomalously large in these magnets, a fact that makes them interesting for practical applications.[4]

In the present paper, we show that the thermodynamic properties of RAA magnets and kinetic phenomena in them should also be very unique. In fact, in the absence of exchange interaction the ground state of each spin is doubly degenerate. The component of the molecular field H transverse to the anisotropy axis splits the ground state by an amount Δ which, as will be shown, is of the order of

$$DS^2(H\sin\theta/DS)^{2S},$$

where D is the single-ion anisotropy constant, S is the spin, and θ is the angle between the local anisotropy axis and the molecular field.1) For the large spins that are characteristic of the heavy rare-earth elements, the splitting Δ is extremely small. For spins whose anisotropy axis is almost perpendicular to the molecular field, the Zeeman splitting $2HS\cos\theta$ is also small.

Thus in RAA magnets there are local excitations whose energy is small not only in comparison with the anisotropy energy DS^2, but also in comparison with the molecular-field energy $HS \approx T_C$. Their density of states is independent of energy down to very small energies of the order of Δ. The properties of these magnets at $T \ll T_C$ should be similar to the properties of the model of two-level systems proposed by Anderson, Halperin, and Varma[5] and also by Phillips[6] to explain the low-temperature anomalies in glasses. But in RAA magnets, in contrast to glasses, the presence of local excitations with a large density of states follows directly from the Hamiltonian of the system.

The specific heat due to transitions between levels for which $HS\cos\theta \approx T$ varies linearly with temperature for $T \approx T_C$, and the temperature variation of the magnetization turns out to be quadratic. Scattering of conduction electrons by such spins leads to a linear temperature variation of the electrical resistivity.

Transitions between weakly split magnetic levels can lead also to other interesting phenomena. For example, resonance absorption of sound of frequency ω by spins for which $2HS\cos\theta = \omega$ is independent of ω over a wide frequency interval and may be larger than the absorption of sound by conduction electrons.

2. SPLITTING OF THE GROUND LEVEL OF THE SPINS, AND THERMODYNAMICS OF THE SYSTEM

RAA magnetic materials are of two types. In intermetallic compounds of heavy rare-earth elements with iron or cobalt, both the ions of the rare-earth elements and those of the transition elements possess a localized magnetic moment. In compounds of rare earths with nickel or nonmagnetic metals, only the rare-earth ions have a magnetic moment.

In the molecular-field approximation, the Hamiltonian

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of the system has the form

$$\mathscr{H}=-\sum_i D(\mathbf{S}_i\boldsymbol{\xi}_i)^2-\sum_{i,m}(\mathbf{H}_{im}\mathbf{S}_{im})+\frac{1}{2}\sum_{i,m}(\mathbf{H}_{im}\langle\mathbf{S}_{im}\rangle), \tag{1}$$

where ξ_i is a unit vector in the direction of the local axis of anisotropy. The molecular field on a spin located at point $\mathbf{r}_{im}$ and belonging to "sublattice" m is

$$\mathbf{H}_{im}=\sum_{jn}V(\mathbf{r}_{im}-\mathbf{r}_{jn})\langle\mathbf{S}_{in}\rangle. \tag{2}$$

Here the angular brackets denote a thermodynamic averaging; $V(\mathbf{r}_{im}-\mathbf{r}_{jn})$ is the Heisenberg exchange-interaction energy of spins located at points $\mathbf{r}_{im}$ and $\mathbf{r}_{jn}$ and belonging to "sublattices" m and n.

The exchange interaction between ions of rare-earth elements is of course small in comparison with the exchange interaction in which iron or cobalt spins take part. Therefore for magnetic materials of the first type, the molecular field on the spins of the rare-earth ions is produced primarily by spins of the other "sublattice," and the iron or cobalt spins are oriented parallel, since their anisotropy energy is small; the moment of the iron or cobalt "sublattice" is antiparallel to the moment of the rare-earth "sublattice."[1]

In compounds of the second type, only the weak interaction between the rare-earth ions leads to ferromagnetic ordering.

In both cases there are spatial fluctuations of the molecular field, caused by fluctuations in the distribution of atoms. Furthermore, in materials of the second type random orientation of the interacting spins also leads to fluctuations of the molecular field. Therefore in these materials the molecular field has a component perpendicular to the magnetization.

If the number z of spins producing the molecular field on a given spin is large, then we may expect that the fluctuations of the molecular field will be small and that the distribution function $W(\mathbf{H})$ will be Gaussian:

$$W(\mathbf{H})=(\pi\overline{H^2})^{-3/2}\exp\{-[H_\perp^2+(H_\parallel-\overline{H})^2]/\overline{H^2}\} \tag{3}$$

for magnetic materials of the second type and

$$W(\mathbf{H})=(\pi\overline{H^2})^{-1/2}\exp\{-(H_\parallel-\overline{H})^2/\overline{H^2}\} \tag{4}$$

for "two-sublattice" magnetic materials; $\overline{H^2}(\overline{H})^{-2}\sim z^{-1}$. The bar denotes a configurational average; $H_\parallel$ and $H_\perp$ are the components of the field along and transverse to the magnetization, respectively.

We consider the system of levels of any rare-earth ion. Its Hamiltonian has the form

$$\mathscr{H}_1=-DS_z^2-\mathbf{HS}, \tag{5}$$

where z is the direction of the anisotropy axis of this spin. In the absence of a molecular field H, the ground state is doubly degenerate, according to the magnetic quantum number $m=\pm S$. If a magnetic field is perpendicular to the z axis, then for classical magnetic moments it only lowers the energy of the ground state by an amount $H^2/4D$. In a quantum-mechanical treatment, splitting of these levels also occurs. It is easy to understand that since the operators S_x and S_y have nonvanishing matrix elements only for states with magnetic quantum numbers different from unity, the splitting of the lower level is $\Delta\sim(H/DS)^{2S}$. For arbitrary mutual orientation of the molecular field and of the anisotropy axis, there occurs, along with the splitting due to the transverse component of the field, a Zeeman splitting. As is shown in Appendix I, the total splitting of the ground-state energy is

$$\varepsilon(H,\theta)=[\Delta^2+4H^2S^2\cos^2\theta]^{1/2}, \tag{6}$$

where

$$\Delta(H)=D\left(\frac{H\sin\theta}{2D}\right)^{2S}\frac{4S}{(2S-1)!}, \tag{7}$$

and where θ is the angle between the local molecular field and the local anisotropy axis. Since we are interested in $\theta\sim\pi/2$, we may replace $\sin\theta$ in (7) by unity.

In the compounds under consideration, $S\gg1$ ($S=6$ for Tb, $S=15/2$ for Dy, $S=8$ for Ho), so that

$$\Delta(H)=\frac{4}{\sqrt{S\pi}}DS^2\left(\frac{eH}{4DS}\right)^{2S},$$

where e is the base of natural logarithms.

Since $2S\gg1$, even for H/DS close to unity the splitting ε is less than the distance DS to the levels with $m=\pm(S-1)$, if $\cos\theta<H/DS$. Therefore the thermodynamics of the system for $T<\min(DS,T_C)$ is determined by transitions between the split sublevels of the ground state. When $T<T_C\sim\overline{H}S$, so that the magnetization is already nearly equal to the saturation magnetization, the specific heat of the magnetic system is

$$C=n\int d^3H\,W(\mathbf{H})\int_0^{\pi/2}d\theta\sin\theta\left(\frac{\varepsilon(H,\theta)}{2T}\right)^2\operatorname{ch}^{-2}\frac{\varepsilon(H,\theta)}{2T}, \tag{8}$$

n is the concentration of the rare-earth atoms.

If $T\gg\Delta$, the specific heat is

$$C=\frac{\pi^2}{12}n\frac{T}{\overline{H}S}\sim n\frac{T}{T_c}. \tag{9}$$

The specific heat (9) is due to spins whose anisotropy axis is almost perpendicular to the molecular field. Since for large S the splitting $\Delta\approx DS^2\exp\{-2S\ln(DS/H)\}$ is exponentially small, the specific heat varies linearly with temperature down to very low temperatures. Numerical calculation shows that the region of linear variation of the specific heat is bounded from above by the temperature $T\approx T_C/3$.

We note that the magnetic specific heat (9) is much larger than the electronic specific heat, since the energy $\overline{H}S$ is small in comparison with the Fermi energy.

At temperatures less than Δ, the specific heat decreases with temperature faster than according to a linear law. If $2S>z$, then the principal contribution to the integral (8) is made by molecular fields such that $\Delta(H)\sim T$. Then

$$C=\frac{\ln2}{2S}H(T)W(H(T)), \tag{10}$$

where $H(T)$ is the value of the molecular field at which

$\Delta(H) = 2T$:

$$H(T)=\frac{4SD}{e}\left(\frac{T\sqrt{S\pi}}{2DS^2}\right)^{1/2S}. \tag{11}$$

But if $2S \ll z$, then the fluctuations of the gap are small in comparison with T, and

$$C \sim \left(\frac{\Delta(\bar{H})}{T}\right)^2 \exp\left(-\frac{\Delta(\bar{H})}{T}\right).$$

The magnetization in an external field H_0 is calculated in a similar fashion. At temperatures $T > \Delta(\bar{H})$ it is

$$\begin{aligned} M(T,H_0) &= n\frac{S}{2} - nS\int_0^1 dx\, x\left(1-\operatorname{th}\frac{Sx(\bar{H}+H_0)}{T}\right) \\ &= \frac{nS}{2}\left(1-\frac{\pi^2}{12}\frac{T^2}{S^2(\bar{H}+H_0)^2}\right). \end{aligned} \tag{12}$$

Hence it is seen that the temperature-dependent part of the susceptibility is proportional to T^2. We note that the spin-wave contribution to $M(T)$ is exponentially small when $T < DS$.

3. THE ELECTRICAL RESISTIVITY

The electrical resistivity of RAA magnetic materials, like their thermodynamic characteristics, should behave anomalously. Supposing that the exchange and nonexchange potentials J and U of interaction of the s electrons with the rare-earth ions are of point nature, we can introduce the relaxation time τ of the s electrons in the usual manner:

$$\tau^{-1}(\varepsilon_k) = \int d^3H\, W(\mathbf{H}) \int_0^{\pi/2} d\theta \sin\theta \sum_{\mathbf{k}'\sigma'} \Phi_{\sigma\sigma'}(\mathbf{k},\mathbf{k}')\frac{1-f_{\mathbf{k}'}}{1-f_{\mathbf{k}}}, \tag{13}$$

where ε_k is the energy of electrons with momentum **k**, f_k is the Fermi distribution function, and $\Phi_{\sigma\sigma'}(\mathbf{k},\mathbf{k}')$ is the probability of scattering of an electron from a state with momentum **k** and spin projection σ ($\sigma = \pm 1$) to a state $\mathbf{k}'$, σ'.

The nonexchange part of the scattering probability is

$$\Phi^U_{\sigma\sigma'}(\mathbf{k},\mathbf{k}') = 2\pi\hbar^{-1}nU^2\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'})\delta_{\sigma\sigma'}, \tag{14}$$

while the exchange part $\Phi^J_{\sigma\sigma'}(\mathbf{k},\mathbf{k}')$ is proportional to the matrix elements of the operator **S**. The value of these matrix elements depends substantially on the ratio between Δ and the Zeeman splitting $2HS\cos\theta$. If $\Delta \gg 2HS \cos\theta$, then in the Hamiltonian (5) we may neglect the longitudinal (along the anisotropy axis) component of the molecular field, and the secular determinant (A.1) becomes symmetric with respect to both diagonals. Therefore the coefficients c^i_m in the expansion of the wave functions ψ_i of the two lower sublevels ($i =$ I, II) in eigenfunctions φ_m of the operator S_z,

$$\psi_i = \sum_{m=-S}^{S} c_m{}^i \varphi_m \tag{15}$$

are symmetric or antisymmetric with respect to replacement of m by $-m$:

$$c_m{}^{\mathrm{I}} = c_{-m}{}^{\mathrm{I}}, \quad c_m{}^{\mathrm{II}} = -c_{-m}{}^{\mathrm{II}}.$$

Hence it follows that

$$\langle\psi_i|S_z|\psi_i\rangle = 0, \quad \langle\psi_i|S^z|\psi_j\rangle = S \quad (i \neq j). \tag{16}$$

The last equality is correct to terms of order H/DS. The matrix elements of the operators $S^{\pm} = S_x \pm S_y$ are zero when $H = 0$. This means that in every case the value of these matrix elements does not exceed H/DS. It can be shown that

$$\langle\psi_i|S^{\pm}|\psi_i\rangle \sim H/DS, \quad \langle\psi_{\mathrm{I}}|S^{\pm}|\psi_{\mathrm{II}}\rangle \sim (H/DS)^{2S-1}.$$

The situation is different when $2HS \cos\theta > \Delta$. In this case the wave functions ψ_{I} and ψ_{II} are composed chiefly of the wave functions φ_S and φ_{-S}, respectively, so that

$$\langle\psi_{\mathrm{I}}|S_z|\psi_{\mathrm{I}}\rangle = -\langle\psi_{\mathrm{II}}|S_z|\psi_{\mathrm{II}}\rangle = S. \tag{17}$$

Since the admixture of states with $|m| < S$ in the wave functions ψ_i is inappreciable, the matrix elements $\langle\psi_{\mathrm{I}}|S_z|\psi_{\mathrm{II}}\rangle$ are small. It is shown in Appendix II that

$$\langle\psi_{\mathrm{I}}|S^z|\psi_{\mathrm{II}}\rangle = S\frac{\Delta}{2HS\cos\theta}, \quad \langle\psi_{\mathrm{I}}|S^{\pm}|\psi_{\mathrm{II}}\rangle \sim \frac{H}{DS}\langle\psi_{\mathrm{I}}|S_z|\psi_{\mathrm{II}}\rangle. \tag{18}$$

It is now clear that the principal role in scattering of electrons is played by scattering without inversion of the electron spin, proportional to matrix elements of the operator S_z. In this case the scattering probability is

$$\begin{aligned} \Phi^J_{\sigma\sigma'}(\mathbf{k}\mathbf{k}') = \frac{2\pi}{\hbar}\delta_{\sigma\sigma'} n \sum_{ii'} N_i\{&-2\sigma JU\langle\psi_i|S_z|\psi_{i'}\rangle \\ &+ J^2\langle\psi_i|S_z|\psi_{i'}\rangle^2\}\delta(\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}'}-\omega_{ii'}). \end{aligned} \tag{19}$$

Here

$$N_i = \exp(-E_i/T)/[\exp(-E_{\mathrm{I}}/T)+\exp(-E_{\mathrm{II}}/T)], \quad \omega_{ii'} = E_{i'} - E_i;$$

E_i is the energy of state i.

On substituting (19) and (14) in (13), expanding in powers of J/U, and calculating the electrical resistivity in the usual way, we get the following expression for the temperature-dependent part of the electrical resistivity, $\Delta\rho(T)$, for $T \gg \Delta(\bar{H})$:

$$\frac{\Delta\rho(T)}{\rho_0} = 16\ln 2\left(\frac{JS}{U}\right)^2\frac{T}{\bar{H}S}, \tag{20}$$

where ρ_0 is the residual resistivity.

The principal contribution to the resistivity (20) comes from elastic scattering by spins for which $2HS\cos\theta \sim T \gg \Delta$. It is due to the diagonal matrix elements of the operator S_z; the nondiagonal matrix element is, according to (18), small in this case.

We notice that $\Delta\rho$ varies linearly with the temperature, and not quadratically as does the moment. This is due to the fact that those spins are important that are almost perpendicular to the molecular field: $\cos\theta \sim T/\bar{H}S \ll 1$. The contribution of each of these spins to the magnetization is small in proportion to the parameter $\cos\theta$, whereas the probability of scattering of conduction electrons by them does not contain this small factor.

At temperatures $T < \Delta(\bar{H})$, the principal role is played by inelastic scattering, due to the matrix element $\langle\psi_{\mathrm{I}}|S_z|\psi_{\mathrm{II}}\rangle$, since the diagonal matrix elements of the operator S_z are small when $HS\cos\theta \sim T \ll \Delta$. In this case the electrical resistivity, like the specific heat, drops with temperature faster than according to a linear law.

4. ABSORPTION OF ULTRASOUND

Transitions between the levels of spins that are almost perpendicular to the molecular field lead to absorption of ultrasound with frequency $\omega \ll HS$. The operator of interaction of a sound wave with the rare-earth ions has the form[7]

$$\mathscr{H}' = \frac{1}{2} \sum_{\alpha,\beta,\gamma,\delta} G_{\alpha\beta\gamma\delta} (S_\alpha S_\beta + S_\beta S_\alpha) e_{\gamma\delta}, \tag{21}$$

where $e_{\gamma\delta}$ is the strain tensor and where G is a tensor that determines the energy of spin-phonon coupling. We may expect that in an amorphous material there will be no relations among the components of the tensor G imposed by the local symmetry.

The absorption of sound by an individual spin is of resonance type. The absorption coefficient γ is determined by the matrix elements of the operators $S_\alpha S_\beta + S_\beta S_\alpha$ between states I and II. Among the matrix elements of these operators for angles $\cos\theta < (\bar{H})^2 (DS)^{-2}$, the largest is the matrix element $\langle \psi_{\mathrm{I}} | S_\alpha S_z + S_z S_\alpha | \psi_{\mathrm{II}} \rangle \equiv L_\alpha(\cos\theta)$ $(\alpha = x, y)$. Calculations similar to those given in the preceding section and in the Appendices showed that

$$L_\alpha(\cos\theta) \approx \begin{cases} \Delta/DS^2 \cos\theta, & \cos\theta > \Delta(\bar{H})/\bar{H}S \\ \bar{H}/DS & \cos\theta < \Delta(\bar{H})/\bar{H}S. \end{cases} \tag{22}$$

In order to calculate γ, it is necessary to calculate the coefficient of acoustical resonance absorption by a single spin[7] and then to average it over orientations of the anisotropy axis and over the molecular-field distribution. For $\omega \ll T$, we have

$$\gamma \approx nF \frac{\omega}{T} \frac{\omega}{\rho v^3} \int d^3H \, W(\mathbf{H}) \int d\cos\theta L_\alpha{}^2(\cos\theta) \delta(\omega - [\Delta^2 + 4H^2S^2\cos^2\theta]^{1/2}). \tag{23}$$

Here ρ is the density, v is the velocity of sound, and $F \sim G^2$. According to Ref. 7, $F \sim 10^{-22}$–10^{-23} erg^2.

In the energy of interaction of a sound wave with localized spins there is in general, along with (21), still another term, due to change of the g factor on strain, and proportional to $H_\alpha S_\beta + H_\beta S_\alpha$.[7] Estimates show that its contribution to the absorption coefficient is of the same order as (23).

It follows from (22) and (23) that in the frequency interval $\Delta(\bar{H}) < \omega < \bar{H}^3/(DS)^2$ the absorption coefficient is

$$\gamma \approx \frac{nF}{\hbar\rho v^3} \left(\frac{\Delta(\bar{H})}{D} \right)^2 \frac{\bar{H}S}{T}. \tag{24}$$

In this frequency range, the absorption coefficient is independent of frequency and inversely proportional to temperature. In the presence of an external magnetic field H_0, the quantity $\bar{H}$ in (24) must be replaced by $\bar{H} + H_0$; that is, an external magnetic field leads to an increase of the absorption. When $ql \ll 1$ (q is the wave vector of the sound, l is the free-path length of the electrons), the absorption of sound by electrons is, as is well known, proportional to ω^2.[8] At sufficiently low frequencies, therefore, the spin absorption exceeds the electronic. Estimates show that for reasonable values of the parameters, the spin absorption exceeds the electronic if $\omega < 10^{10} - 10^9$ GHz.

When $\omega < \Delta(\bar{H})$, the nature of the variation of γ with frequency is determined by the standard deviation of the gap distribution, δ. If $\delta \ll \Delta(\bar{H})$, the absorption coefficient has a threshold singularity; that is,

$$\gamma(\omega) \sim [\Delta(\bar{H})/(\omega - \Delta(\bar{H}))]^{1/2}, \quad \omega - \Delta(\bar{H}) > \delta,$$

while for $|\omega - \Delta(\bar{H})|$ the absorption coefficient differs from (24) by a factor $(\Delta(\bar{H})/\delta)^{1/2} \gg 1$. On further decrease of frequency, the absorption coefficient drops exponentially fast.

If, however, the standard deviation of $\Delta(H)$ is of the same order as $\Delta(\bar{H})$, then there are no threshold singularities, and at frequencies less than $\Delta(\bar{H})$ the absorption coefficient decreases smoothly.

5. CONCLUSION

In the Harris-Plischke-Zuckermann model that we have adopted [the Hamiltonian (1)], the simplest form of the anisotropy energy was chosen. We shall consider to what consequences a more realistic model may lead.

RAA magnetic materials have a structure of the random close packing type[9,10]; therefore the environment of each rare-earth ion in general does not have axial symmetry, so that the anisotropy energy must be of the form[10]

$$\mathscr{H}_{\text{anis}} = -DS_z{}^2 - D_1S_x{}^2, \quad D > D_1.$$

The term $D_1S_x^2$ leads to splitting of the ground state by the amount $\Delta_1 \sim DS^2(D_1/D)^S$. If $D_1 > D(H/DS)^2$, then $\Delta_1 > \Delta$, and the matrix elements of transition between states I and II increase accordingly. At the same time, since at large spins Δ_1 is proportional to a high power of D_1/D, Δ_1 is much smaller than T_C, so that the results obtained above remain valid with replacement of Δ by Δ_1. The same consequences may also result from terms in the anisotropy energy that are of higher order in the spin operators.

In consequence of the amorphousness of the structure of RAA magnetic materials, there are fluctuations not only of the molecular field but also of the anisotropy constant D, so that in calculating physical quantities one must in general average over the distribution of D. Such averaging obviously does not change the results qualitatively but requires replacement of D by some effective value $\bar{D}$.

The possibility has recently been discussed that in "single-sublattice" RAA magnets only the spin-glass state is stable.[11] Since our results are due to local spin flips, they are not dependent on the type of long-range magnetic order.

Finally, we note the following. Because RAA magnetic materials are isotropic on the average (macroscopically), the ground state is degenerate with respect to the direction of the total moment. A consequence of this should be the existence of a Goldstone mode (perhaps diffusional), which may also make a contribution to the thermodynamics.

APPENDIX I

The secular equation for the Hamiltonian (5) has the form

$$\begin{vmatrix} -E-HS\cos\theta & a_{12} & 0 & \dots & 0 & 0 \\ a_{12} & a_{22}-E & a_{23} & & 0 & 0 \\ 0 & a_{23} & a_{33}-E & & 0 & 0 \\ 0 & 0 & a_{34} & & 0 & 0 \\ \dots & \dots & \dots & & \dots & \dots \\ 0 & 0 & 0 & & a_{2S,2S}-E & a_{12} \\ 0 & 0 & 0 & & a_{12} & -E+HS\cos\theta \end{vmatrix} = 0. \tag{A.1}$$

Here E is the energy, measured from $-DS^2$;

$$\begin{gathered} a_{l,l+1}=a_{l+1,l}={}^1/_2[l(2S-l+1)]^{1/2}H\sin\theta, \\ a_{ll}=D(l-1)(2S-l+1)-H(S-l+1)\cos\theta, \\ 1\leqslant l\leqslant 2S+1. \end{gathered} \tag{A.2}$$

The determinant (A.1) is symmetric about the principal diagonal.

We denote by $B_{ij}(E)$ the determinant obtained from (A.1) by deletion of the $i-1$ top rows and left columns and of the $j-1$ bottom rows and right columns. The determinant B_{ij} satisfies the recurrence relations

$$\begin{gathered} B_{ij}=a_{ii}B_{i+1,j}-a_{i,i+1}^2B_{i+2,j}, \quad i,j<S, \\ B_{ij}=a_{2(S+1)-j,2(S+1)-j}B_{i,j+1}-a_{j,j+1}B_{i,j+2}. \end{gathered} \tag{A.3}$$

With the aid of (A.3), we transform equation (A.1) to the form

$$\begin{gathered} E^2B_{22}(E)+Ea_{12}^2(B_{23}(E)+B_{32}(E))+a_{12}^4B_{33}(E) \\ -H^2S^2\cos^2\theta B_{22}(E)+a_{12}^2HS\cos\theta(B_{23}-B_{32})=0. \end{gathered} \tag{A.4}$$

Hence

$$\begin{gathered} E=-\frac{a_{12}^2}{2B_{22}}(B_{23}+B_{32})\pm\frac{1}{2}\Big[\Delta^2+4H^2S^2\cos^2\theta \\ -\frac{4a_{12}^2}{B_{22}}(B_{23}-B_{32})HS\cos\theta\Big]^{1/2}, \end{gathered} \tag{A.5}$$

where

$$\Delta^2=\frac{a_{12}^4}{B_{22}^2(E)}\{(B_{23}(E)+B_{32}(E))^2-4B_{22}(E)B_{33}(E)\}. \tag{A.6}$$

On transforming the determinants in the numerator of (A.6) by use of the recurrence relations (A.3), we get

$$\begin{gathered} \Delta^2=a_{12}^4B_{22}^{-2}\{a_{23}^4[(B_{34}+B_{43})^2-4B_{33}B_{44}]+4a_{23}^2B_{33} \\ \times(B_{43}-B_{34})H(S-1)\cos\theta+4B_{33}^2H^2(S-1)^2\cos^2\theta\}. \end{gathered} \tag{A.7}$$

By continuing this procedure, one can easily show that, to the lowest order in H/DS,

$$\Delta=2\frac{a_{12}^2a_{23}^2\dots}{a_{22}a_{33}a_{44}\dots}. \tag{A.8}$$

In the same approximation, the last term under the radical sign in (A.5) is a small correction to $H^2S^2\cos^2\theta$.

By using formulas (A.2) and supposing that $\sin\theta\approx 1$, we obtain from (A.5) and (A.8) the expressions (6) and (7) of the main text for $\varepsilon=E_1-E_2$ and for Δ. According to (A.5) and (A.2), the shift of the levels is $-(H^2/4D)2S/(2S-1)$.

APPENDIX II

We shall calculate the matrix element $\langle\psi_{\rm I}|S_z|\psi_{\rm II}\rangle$ for $HS\cos\theta\gg\Delta$. According to (15), we have

$$\langle\psi_{\rm I}|S_z|\psi_{\rm II}\rangle=\sum_{m=S}^{-S}mc_m^{\rm I}c_m^{\rm II}. \tag{A.9}$$

The coefficients c_m^i are determined by the system of equations

$$\sum_{k=1}^{2S+1}c_{S+1-k}^ia_{nk}(E_i)=0 \tag{A.10}$$

with the determinant (A.1). All the coefficients in (A.10) can be expressed in terms of c_S^i:

$$c_{S+1-k}^i=c_S^i\frac{B_{1,2S+3-k}}{a_{12}a_{23}\dots a_{k-1,k}},\quad k=2,3,\dots,2S+1. \tag{A.11}$$

We shall find the coefficient c_{-S}^i:

$$c_{-S}^i=c_S^i\frac{B_{12}}{a_{12}a_{23}\dots a_{2S,2S+1}}. \tag{A.12}$$

By using the recurrence relation (A.3) and the expressions (A.5) and (A.8), we get from (A.12) for $j=\mathrm{II}$:

$$c_{-S}^{\rm II}=-c_S^{\rm II}\cdot 4HS\cos\theta/\Delta. \tag{A.13}$$

Similarly for $i=\mathrm{I}$, one gets

$$c_S^{\rm I}=c_{-S}^{\rm I}\cdot 4HS\cos\theta/\Delta. \tag{A.14}$$

With the aid of (A.11) one can show that the coefficients c_m^i decrease with decrease of $|m|$. Therefore the principal contribution to the sum (A.9) comes from terms with $m=\pm S$. By taking into account the normalization condition

$$\sum_{m=S}^{-S}(c_m^i)^2=1,$$

we finally get

$$\langle\psi_{\rm I}|S_z|\psi_{\rm II}\rangle=S\frac{\Delta}{2HS\cos\theta}. \tag{A.15}$$

Consider the matrix element

$$\langle\psi_{\rm I}|S^+|\psi_{\rm II}\rangle=\sum_m[(S+m)(S-m+1)]^{1/2}c_m^{\rm I}c_{m-1}^{\rm II}. \tag{A.16}$$

Since c_m^i decreases with decrease of $|m|$, the principal contribution to (A.16) comes from the term with $m=S$, and according to (A.11) and (A.13)–(A.15)

$$\langle\psi_{\rm I}|S^+|\psi_{\rm I}\rangle\sim\frac{H}{DS}\langle\psi_{\rm I}|S_z|\psi_{\rm II}\rangle. \tag{A.17}$$

The matrix element $\langle\psi_{\rm I}|S^-|\psi_{\rm II}\rangle$ is calculated similarly.

Note added in proof (August 23, 1978). As became known to us recently, a linear variation of the specific heat with temperature, caused by local magnetic excitations, has been observed in the amorphous compound DyCu [J. M. D. Coey, J. Appl. Phys. **49**, 1636 (1978)].

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[1] The field H and the temperature T are measured in energy units.

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Dragging of electrons by sound in metals

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A sound wave attenuating in a metal transfers its momentum to the conduction electrons, thus dragging the electrons. This produces in the sample an acoustoelectric voltage v_{ph} and an acoustomagnetic field H_{ph} if the sound flux occupies respectively the entire or part of the sample cross section. These effects were investigated in tin, aluminum, bismuth, and gallium in the interval 4.2–1.2 K. It is shown that dragging of electrons by sound does not depend on temperature; tin has a giant anisotropy of the acoustoelectric voltage v_{ph}, probably as a result of singularities of the Fermi surface. Nonlinear effects connected with the acoustomagnetic field H_{ph} occur in gallium when the sound intensity is increased.

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INTRODUCTION

The damping of elastic (acoustic) waves in metals at low temperatures is due mainly to their interaction with the conduction electrons. The energy and momentum of the wave are first transferred to the electrons, and are then dissipated when already in the electron system. In this case the sound drags the conduction electrons. A propagating sound wave excites in a conductor a current and an electric field. The possible existence of this effect was first pointed out by Parmenter,[1] who called it acoustoelectric. Although it soon became clear (see, e.g., Ref. 2) that Parmenter's theoretical calculations were in error, the very existence of electron dragging by the sound was subject to no doubt.

When the wave loses an energy Q as a result of interaction with the conduction electrons, the momentum of the elastic wave Q/u (u is the speed of sound) is transferred to the electrons. As a result, in the simplest case, a current flows in the conductor, given by

$$I=e^*\tau Q/m^*u, \tag{1}$$

where e^* and m^* are the charge and mass of the carriers, and τ is the relaxation time in the conduction-electron system. The direction of the current depends on whether the charge carriers are electrons or holes.

If the sample circuit is open, then an electric voltage is produced along the conductor

$$E_{ph}=Q/e^*uN, \tag{2}$$

where N is the charge density. It is seen from (2) that the acoustoelectric voltage is larger the smaller the charge density. This is probably why until recently this effect was investigated only in semiconductors[3] or in semimetals.[4,5] In typical metals the expected effect is too small to be detected by the existing measurement methods.

Indeed, assume that optimal conditions are realized in the experiment, and the voltage produced in the sample is due to absorption of the entire energy of the sound wave. (For this purpose it is necessary that the distance between the sample points between which the voltage $V_{ph}=\int E_{ph}\,dy$, is measured be so large that the sound wave is completely damped between them.) Then

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