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Directionally Independent Energy Gap Formation Due to the Hyperfine Interaction

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We study energy gap formation at the level-crossing point due to the hyperfine interaction. In contrast to the energy gap induced by the Dzyaloshinskii-Moriya interaction, the gap induced by the hyperfine interaction is independent of the direction of the magnetic field. We also study the dynamics of the magnetization under a time dependent field that changes at a constant rate.

§1. Introduction

Recently, adiabatic magnetic transitions in nanoscale magnets have attracted a great deal of interest. Among many interesting problems in this context is the search for the origin of the transition or, in other words, to determine how an energy gap is formed at the level-crossing point.[1)] The Dzyaloshinskii-Moriya interaction (DMI) has been proposed as one of the possible origins of this energy gap.[2)−5)] Very recently, however, it was shown that, on a triangular lattice with three spins and for a 15-spin model of the V_{15} molecule, the gap formed due to the DMI is inevitably dependent on the magnetic field direction.[6)] Thus, for some directions of the applied magnetic field, an adiabatic transition cannot be realized. Even if the extended DMI[7)] or an interaction taking the form of the scalar product of three spins is considered, the directional dependence of the gap cannot be removed.

In this paper, we study the formation of an energy gap due to the hyperfine interaction. Because the magnetic moments of the electron and nuclear spins are different, the Zeeman interaction does not commute with the interaction of the Heisenberg model. Therefore, quantum mixing, or an adiabatic change of the magnetization, can be realized when the magnitude of the magnetic field is changed smoothly. The gap caused by the hyperfine interaction is independent of the direction of the applied magnetic field. The behavior we study is of particular interest because quantum mixing due to the hyperfine interaction has been observed experimentally in a single crystalline 0.2% holmium doped LiYF by Giraud et al.[8)]

To this time, the hyperfine interaction has been treated as noise that broadens the energy levels of electron spin systems. In this paper, we study gap formation due

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to the hyperfine interaction in two simple systems: a dimer consisting of one electron and one nuclear spin, and a triangular system consisting of three such dimers. For both systems, we investigate how the magnetization changes when the magnitude of the magnetic field changes at a finite rate.

If we consider coupling to nuclear spins, the energy levels no longer consist simply of the levels of the electron spin system, and the system possesses sets of many closely-spaced levels. When considering the crossing of such a set of energy levels, we cannot simply estimate the probability of the adiabatic change of a state using the Landau-Zener-Stückelberg (LZS) formula, [1),9)]

$$P = 1 - \exp\left(-\frac{\pi(\Delta E)^2}{2\hbar v}\right), \tag{1·1}$$

where ΔE is the energy gap at the crossing point and v is the rate at which the Zeeman energy changes. Hereafter, we use units in which $\hbar = 1$. For the triangular system, we study how the rate of the adiabatic change of the magnetization, starting from the ground state, depends on the rate of change of the magnetic field strength. We also study the dynamics of the magnetization of a thermally populated system. The dependence of the magnetization on the rate of change of the field strength is analyzed using an effective LZS formula.

§2. Gap formation due to the hyperfine interaction

First, we consider the simplest case of a dimer consisting of one electron spin $\boldsymbol{S}$ and one nuclear spin $\boldsymbol{I}$. The Hamiltonian for this system is given by

$$\mathcal{H} = a\boldsymbol{S}\cdot\boldsymbol{I} - (g\mu_{\rm B}\boldsymbol{S} + g'\mu_{\rm N}\boldsymbol{I})\cdot\boldsymbol{H}, \tag{2·1}$$

where a denotes the strength of the hyperfine interaction between spins $\boldsymbol{S}$ and $\boldsymbol{I}$, $\mu_{\rm B}$ and $\mu_{\rm N}$ represent the magnetic moments and g and g' the gyromagnetic ratios of $\boldsymbol{S}$ and $\boldsymbol{I}$, respectively, and $\boldsymbol{H}$ denotes the external magnetic field. We assume $|\boldsymbol{I}| = |\boldsymbol{S}| = 1/2$. Because $g\mu_{\rm B} \gg g'\mu_{\rm N}$, we assume $g'\mu_{\rm N} \simeq 0$. Then, taking the magnetic field along to be the z-direction, Eq. (2·1) finally reduces to

$$\mathcal{H} = a\boldsymbol{S}\cdot\boldsymbol{I} - hS_z, \tag{2·2}$$

where h denotes the strength of the external magnetic field. The energy levels of this Hamiltonian read

$$\begin{aligned} E_1 &= -\tfrac{1}{4}a - \tfrac{1}{2}\sqrt{a^2+h^2}, \\ E_2 &= \tfrac{1}{4}a + \tfrac{1}{2}h, \\ E_3 &= -\tfrac{1}{4}a + \tfrac{1}{2}\sqrt{a^2+h^2}, \\ E_4 &= \tfrac{1}{4}a - \tfrac{1}{2}h. \end{aligned} \tag{2·3}$$

For $h \approx 0$, the energy level diagram is depicted in Fig. 1. Here and hereafter, we set $a = 1$ and $g\mu_{\rm B} = 1$, which determine the units employed here. Two types of level crossing structures are seen: Levels E_2 and E_4 simply cross, while levels E_1 and E_3

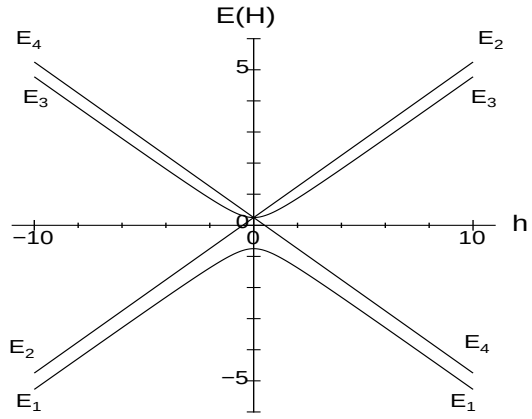

Fig. 1. Energy level diagram of the Hamiltonian (2·2) with $a = 1$ and $g\mu_{\mathrm{B}} = 1$.

form an avoided level crossing structure. Note that the energy level diagram does not depend on the direction of the H-field.

If the temperature is sufficiently low and if mainly the lowest level E_1 is occupied, simple adiabatic motion is expected. Such adiabatic motion is realized without introducing an asymmetric interaction, such as the DMI, which is necessary for gap formation in an electron spin system with time-reversal symmetry.[4)] The hyperfine interaction does not break the time-reversal symmetry and does not introduce geometrical anisotropy. However, because of the difference between the magnetic moments, even for a uniform field, the Hamiltonian does not commute with the Zeeman energy, and this leads to the opening of a gap. Thus, the hyperfine interaction provides a mechanism for the formation of a gap that does not depend on the direction of the field.

Let us now consider the change in magnetization at finite temperature when the strength of the field is changed continuously from a large negative value to a large positive value. For negative values of h, the energy difference between levels E_1 and E_2 is of the same order of magnitude as the energy gap at $h = 0$, which is caused by the hyperfine interaction a. Thus, at a temperature of order a, the energy levels E_1 and E_2 are almost equally occupied, if we assume that the system started from an equilibrium distribution. When the strength of the magnetic field is slowly changed from negative to positive values, the population p_1 of level E_1 remains unchanged, and the magnetization of the state changes smoothly from $-1/2$ to $1/2$. The population p_2 of level E_2 also remains unchanged. In this case, the magnetization of the state does not change adiabatically and remains $-1/2$ for any rate of change of the field strength, as long as we consider only quantum mechanical motion. When the sign of the field changes, the state becomes metastable, and thermal noise can cause relaxation of the metastable magnetization from $-1/2$ to $1/2$. Then the state changes from level E_2 to level E_4 or E_1. Although this relaxation process depends on how the system is coupled to the thermal bath, the relaxation always occurs after the field passes the crossing point at $h = 0$.

In the whole process, half of the population with $m = -1/2$ changes adiabatically to $m = 1/2$, and the other half relaxes through a thermal process to $m = 1/2$. Therefore, we can imagine that the magnetization changes from $-1/2$ to 0 just after the field changes sign, and this is followed by slow relaxation from 0 to 1/2. In this process, we expect a plateau in the magnetization just after the field changes sign. This effect is reminiscent of the magnetic plateau phenomena observed in magnetic iron clusters [10),11)] and in the V_{15} molecule, which appear as the result of a continuously changing magnetic field. [2)] These phenomena can be understood from the point of view of the magnetic Föhn effect (MFE); [12)−14)] that is, a small heat inflow that occurs during the adiabatic change at the crossing region causes a temperature increase after the crossing. In this mechanism, when the sweeping velocity becomes slow, more heat flows in, and the height of the magnetization plateau decreases.

It is important to note that the system studied above has two different types of crossings. A fast rate of change of the magnetic field causes a large deviation of the population of E_2 from its value in the equilibrium state. This is due to the non-adiabatic LZS transition, not the thermal effect (MFE).

A change of the magnetization due to a combination of both mechanisms, i.e., MFE for the population of E_1 and the LZS transition for the population of E_2, generally leads to a complicated dependence of the magnetization on the temperature and the rate of change of the magnetic field strength.

§3. Triangular system

We consider the case of three electron spins on a triangular lattice in the presence of a DMI. The Hamiltonian for this system reads

$$\mathcal{H} = J(\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1) + \sum_{i>j} \boldsymbol{D}_{ij} \cdot (\boldsymbol{S}_i \times \boldsymbol{S}_j). \tag{3·1}$$

The antiferromagnetic Heisenberg interactions of strength J generate doubly degenerate doublets, and the DMI can cause an energy gap. [2)−4)] Taking into account the symmetry of the lattice (C_3), the vectors of the DMI satisfy the relations

$$D_{12}^z = D_{23}^z = D_{31}^z \tag{3·2}$$

and

$$\begin{pmatrix} D_{12}^x \\ D_{12}^y \end{pmatrix} = \begin{pmatrix} -1/2 & \sqrt{3}/2 \\ -\sqrt{3}/2 & -1/2 \end{pmatrix} \begin{pmatrix} D_{23}^x \\ D_{23}^y \end{pmatrix} = \begin{pmatrix} -1/2 & \sqrt{3}/2 \\ -\sqrt{3}/2 & -1/2 \end{pmatrix}^2 \begin{pmatrix} D_{31}^x \\ D_{31}^y \end{pmatrix}. \tag{3·3}$$

If a magnetic field is applied along the x-direction, the system has degenerate avoided level crossings. [3)] However, it has been found that when the above relations hold among the DM vectors, the energy levels do not form an avoided level crossing structure when the field is applied along the z-direction. [6)] A similar dependence of the energy gap on the direction of the magnetic field has been found in the 15-spin

system modeling the V_{15} molecule.[6] This type of dependence of the gap on the magnetic field direction seems to be intrinsic to the DMI.

Now, we consider a system with the hyperfine interaction instead of DMI. As shown in the previous section, the hyperfine interaction can provide a gap that is independent of the field direction. The hyperfine interaction provides a mechanism to generate energy gaps that differs from that of DMI.

We consider a model of three dimers consisting of an electron spin $\boldsymbol{S}$ and a nuclear spin $\boldsymbol{I}$ [see Fig. 2(a)]. The Hamiltonian is given by

$$\mathcal{H} = J(\boldsymbol{S}_1 \cdot \boldsymbol{S}_2 + \boldsymbol{S}_2 \cdot \boldsymbol{S}_3 + \boldsymbol{S}_3 \cdot \boldsymbol{S}_1) + a(\boldsymbol{S}_1 \cdot \boldsymbol{I}_1 + \boldsymbol{S}_2 \cdot \boldsymbol{I}_2 + \boldsymbol{S}_3 \cdot \boldsymbol{I}_3) - \sum_{i=1}^{3} (g\mu_{\mathrm{B}} \boldsymbol{S}_i + g'\mu_{\mathrm{N}} \boldsymbol{I}_i) \cdot \boldsymbol{H}. \tag{3·4}$$

We set $g\mu_{\mathrm{B}} = 1$ and $g'\mu_{\mathrm{N}} = 0.001$. Each of the 8 energy levels of the electron spin interactions is split into 8 levels, due to the hyperfine interaction. Hence, in total there are 64 energy levels. In order to make the effect of the hyperfine interaction clearly visible, we use a rather large value for its strength, a, choosing $a = 1$ and $J = 10$. The energy level diagram of the model given in (3·4) is depicted in Fig. 2(b).

In the figure, two degenerate doublets ($S = 1/2$) and quartet ($S = 3/2$) structures of the electron spin interaction are seen to persist, although each level of the electron interaction is broadened, due to the degree of freedom of the nuclear spins.

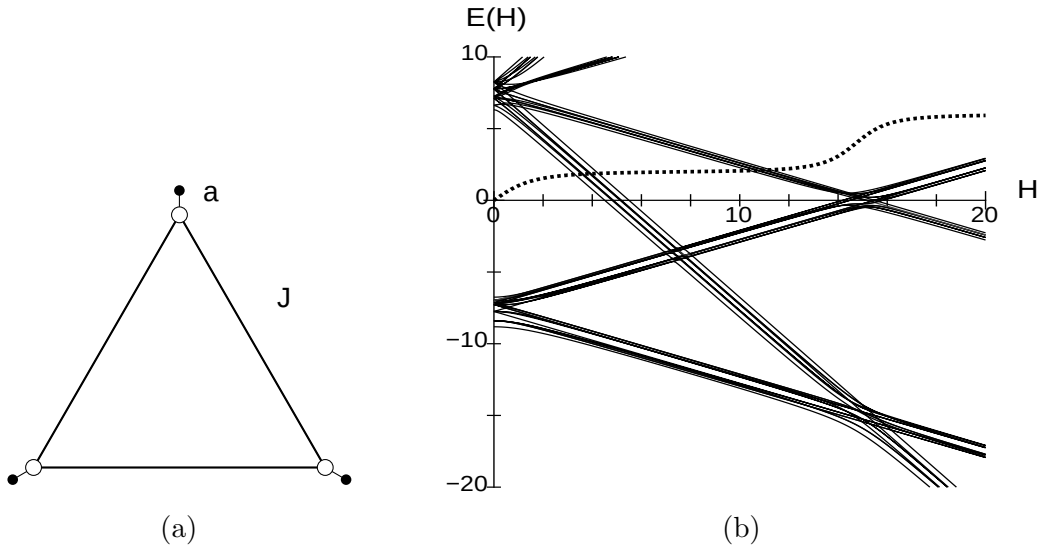

Fig. 2. (a) A system of three dimers, each consisting of one electron (unfilled circle) and one nuclear (filled circle) spin on a triangular lattice. The electron spins interact with each other via an antiferromagnetic Heisenberg interaction of strength J. The electron and nuclear spins interact via the hyperfine interaction a. (b) The energy level diagram of the Hamiltonian given in (3·4) with $J = 10$, $a = 1$, $g\mu_{\mathrm{B}} = 1$ and $g\mu_{\mathrm{N}} = 0.001$. The dotted curve represents the magnetization of the ground state, which is multiplied by a factor of 4 for the sake of clarity.

The width of the splitting is $O(a)$. Each group of levels in the figure consists of 8 levels. In the present model, the system has two-fold degenerate doublets. Thus, the group of levels of magnetization $M = 1/2$, i.e., the group with the lowest energy for $H = 4$, consists of 16 levels. At $H = 0$, the groups with $M = \pm 1/2$ merge, and as a result, 32 levels form a nest of levels around $E = -8J$. Instead of the simple level crossing structure seen in systems with only electron spins, this system exhibits crossings of groups of levels. At each crossing, there is complicated level structure.

It should be noted that the ground state level does not cross any other level. Hence, the magnetization of the ground state level can be changed adiabatically from $-3/2$ to $3/2$ by slowly changing the magnetic field strength from negative to positive values. This contrasts with the behavior that is observed for the model given in (3·1).[6] In that model, at least two levels have to cross at $H = 0$ to form a Kramers doublet.[4] Thus, the ground state for negative values of the magnetic field becomes the second lowest state after the field has changed sign. The magnetization, starting from $-3/2$, cannot be caused to reach $3/2$ adiabatically by slowly changing the magnetic field. This is one of the peculiar consequences of the energy structure of a triangular antiferromagnetic system with DMI.[4]

§4. Dynamical properties of a triangular antiferromagnet with the hyperfine interaction

As the number of nuclear spins increases, the energy levels of the electron spin broaden. Usually, this broadening is considered to be an effect of noise. In what follows, we study the quantum mechanical dynamics of such broadened levels and investigate whether the picture of the LZS transition still holds. For this purpose, we study the dynamical properties of the triangular system (3·4).

First, we study the magnetization process in a continuously changing magnetic field. Choosing the initial field strength to be $H = -20$, we obtain all the eigenstates of the model with this field strength and label them $|i\rangle$ $(i = 1, 2, \cdots, 64)$. For each of these 64 states, we solve the time-dependent Schrödinger equation

$$i\frac{\partial}{\partial t}|\Psi(t)\rangle = \mathcal{H}(t)|\Psi(t)\rangle, \tag{4·1}$$

where $\mathcal{H}(t)$ is the Hamiltonian (3·4) and the three components of the time-dependent field read

$$\boldsymbol{H} = (0, 0, -20 + vt). \tag{4·2}$$

For each state $\{|i(t)\rangle\}$, we calculate the magnetization

$$m_i(t) = \langle i(t)| \sum_{j=1}^{3} (g\mu_{\mathrm{B}} S^z{}_i + g'\mu_{\mathrm{N}} I^z{}_j)|i(t)\rangle. \tag{4·3}$$

Here, we set $a = 1$, $J = 10$ and $v = 0.5$. In Fig. 3, we depict $\{m_i(t)\}$ for $i = 1$ – 64. Various types of evolution of the magnetization can be seen. For the chosen parameter set, the magnetization starting from the ground state behaves almost

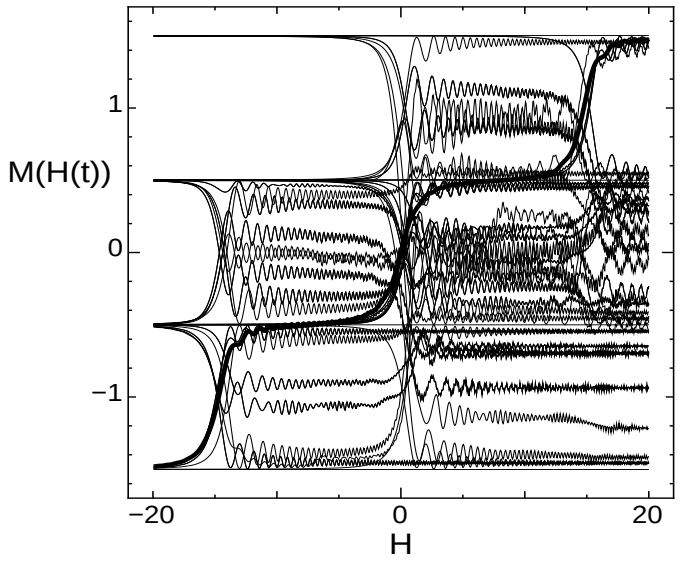

Fig. 3. Time evolution of the magnetization for each of the 64 eigenstates of the Hamiltonian (3·4), with $a = 1$, $J = 10$ and $v = 0.5$. The bold curve represents the magnetization change starting from the ground state.

adiabatically when the field is changed from $-H_0$ $(= -20)$ to H_0 (see the bold curve).

Next, we study the dependence of the adiabatic change on the rate of change of the field strength. We concentrate on the change of the magnetization around $H = 0$. Here, we consider the magnetic field to change at a rate v from $-H_0$ to H_0 and choose $H_0 = 5.0$.

Because we are interested in the dependence of the transition on v, we consider the range of values of v from that for an almost diabatic transition (i.e. no change of spin configuration) to that for an almost adiabatic transition. Although the LZS formula (1·1) holds for any combination of parameters, the time over which H increases, H_0/v, should be larger than the precession time, $\sim O(2\pi/H)$, to estimate the transition probability. Actually, in Fig. 3, we see the oscillation due to the precession clearly. We now consider the case $a = 0.1$ to make the energy gap small. This is more convenient to study the effect of a non-adiabatic transition.

If we choose the unit of energy J to be 1K $(=1k_{\rm B} = 1.38 \times 10^{-23}{\rm J})$, taking into account the values $\hbar = 1.05 \times 10^{-34}{\rm Js}$ and $\mu_{\rm B} = 9.27 \times 10^{-24}$ J/T, the velocity $v = 0.5$ corresponds approximately to $v_0 = 1.0 \times 10^{11}[{\rm T/s}]$.*) Let the hyperfine interaction be of order 10^{-4}K. Then, a should be taken to be 10^{-4}, which makes the energy gap smaller by a factor of approximately 10^{-4}. In this case, v must take a

*) Explicitly, we have

$$v = 0.5 = \frac{v_0 \mu_{\rm B} \hbar}{J^2} = \frac{v_0 {\rm T/s} 9.27 \times 10^{-24}{\rm J/T} \times 1.05 \times 10^{-34}{\rm Js}}{(1.38 \times 10^{-23}{\rm J})^2}.$$

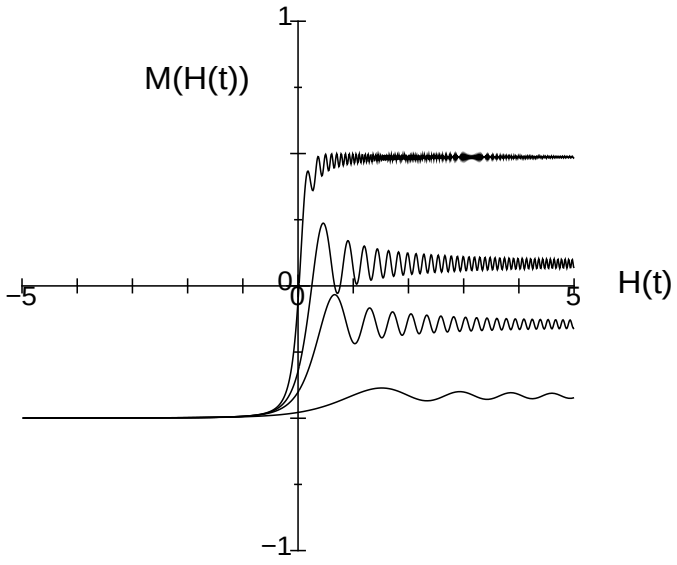

Fig. 4. Time evolution of the magnetization starting from the ground state for $v = 0.01$, 0.05, 0.1 and 0.5 (from top to bottom) and $a = 0.1$, $J = 10$.

value in the range 0.01×10^{-8} – 0.5×10^{-8} in order to have the same change of the transition probabilities as listed in Table I. This range corresponds $v_0 = 0.02 \times 10^3$ – 1.0×10^3[T/s].

We estimate the amount of adiabatic change by

$$P = |\langle \mathrm{G}(H_0)|\Psi(t_\mathrm{f})\rangle|^2 , \tag{4·4}$$

where $t_\mathrm{f} = 2H_0/v$, and $G(H_0)$ denotes the ground state at $H = H_0$. Typical time evolutions of the magnetization starting from the ground state are depicted in Fig. 4 for various values of v. Table I displays the dependence of P on v. The effective energy gap ΔE can be estimated from P by making use of the LZS formula,[1),9)]

$$\Delta E = \sqrt{-2v \log(1-P)/\pi}. \tag{4·5}$$

Values of ΔE are also given in Table I.

Table I. Probability P of adiabatic change, (4·4), and the effective energy gap ΔE, estimated from the LZS formula (4·5) for various values of the rate of changing of the field strength, v.

v	P	ΔE
0.500	0.08371	0.16681
0.250	0.16067	0.16697
0.100	0.35481	0.16702
0.050	0.58378	0.16704
0.025	0.82678	0.16704
0.010	0.98751	0.16704

For $a = 0.1$, the energy gap between the ground state and the 32nd level is 0.20067, which is the maximum width of the lowest energy band at $H = 0$, as mentioned in §2. We thus find that the LZS formula works well with an effective energy gap ($\Delta E \approx 0.167$), although the energy level structure is rather complicated.

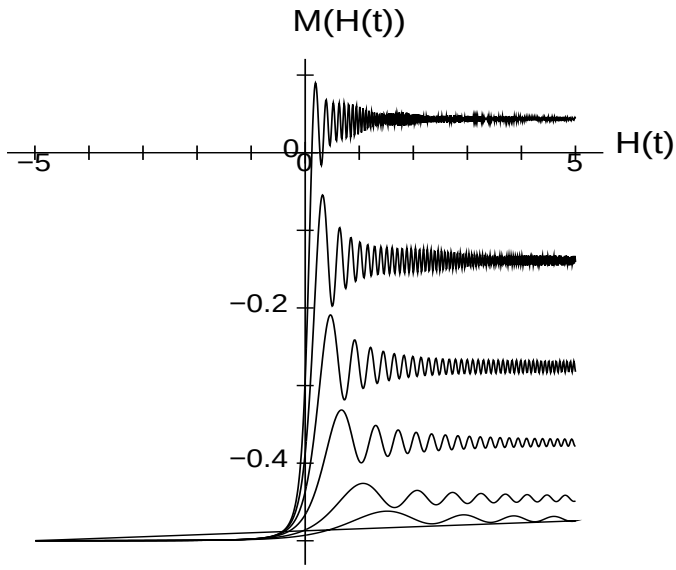

Fig. 5. Time evolution of the magnetization starting from an equilibrium state at temperature $T = 0.2$. Here $v = 0.01$, 0.025, 0.05, 0.1, 0.25, 0.5 and 1.0 (from top to bottom).

If we assume that initially the states are distributed according to the canonical distribution, we can calculate the magnetization as

$$\langle m(t) \rangle = \frac{\sum_{i=1}^{64} m_i(t) e^{-\beta E_i}}{\sum_{i=1}^{64} e^{-\beta E_i}}, \tag{4·6}$$

where $\beta = 1/k_{\mathrm{B}}T$, k_{B} denotes Boltzmann's constant, and T is the temperature. The dependence of $\langle m(t) \rangle$ on v is shown in Fig. 5. The effective transition probability P_{eff} is estimated using the thermal average of the overlap function as

$$P_{\mathrm{eff}} \equiv \langle P \rangle = \sum_{i=1}^{16} \left| \langle i(t_{\mathrm{f}}) | \Psi(t_{\mathrm{f}}) \rangle \right|^2, \tag{4·7}$$

where the states $i = 1, \cdots, 16$ are those in the group for which $m_i \simeq 1/2$. We regard this probability P_{eff} as the rate of adiabatic change between the groups of levels with $m = \pm 1/2$. The transition probability P_{eff} and the effective gap estimated by (4·5) are listed in Tables II and III, respectively.

We conclude that when the temperature is of order $a = 0.1$, the LZS formula seems to work well, and the estimated effective gap is equal to approximately half of the maximum width of the lowest band at $H = 0$. Although more work is necessary to provide more quantitatively precise arguments, we conclude that the LZS-type dependence of the probability of the adiabatic change on the rate of change of the magnetic field provides a good approximation and that we can estimate the size of the energy gap from this dependence.

Table II. Transition probability $P_{\rm eff}$, (4·7), for various rates of change of the field strength v and temperatures T.

v	$T=1.0$	$T=0.5$	$T=0.3$	$T=0.2$	$T=0.1$	$T=0.05$	$T=0.01$
0.50000	0.03147	0.02555	0.02603	0.02666	0.02850	0.03165	0.03839
0.25000	0.05620	0.05113	0.05229	0.05378	0.05811	0.06566	0.08412
0.10000	0.12773	0.12556	0.12932	0.13404	0.14788	0.17271	0.24280
0.05000	0.20470	0.20349	0.20709	0.21156	0.22429	0.24557	0.28213
0.02500	0.34472	0.34911	0.35769	0.36834	0.39930	0.45362	0.58738
0.01000	0.51469	0.52541	0.53942	0.55681	0.60754	0.69735	0.93007

Table III. Effective gap $\Delta E_{\rm eff}$, estimated using the LZS formula, (4·5), for various rates of change of the field strength v and temperatures T.

v	$T=1.0$	$T=0.5$	$T=0.3$	$T=0.2$	$T=0.1$	$T=0.05$	$T=0.01$
0.50000	0.10089	0.09077	0.09162	0.09275	0.09594	0.10117	0.11163
0.25000	0.09595	0.09140	0.09246	0.09380	0.09761	0.10397	0.11826
0.10000	0.09327	0.09242	0.09389	0.09572	0.10093	0.10986	0.13306
0.05000	0.08539	0.08510	0.08594	0.08698	0.08991	0.09471	0.10272
0.02500	0.08202	0.08267	0.08394	0.08551	0.09006	0.09808	0.11870
0.01000	0.06784	0.06888	0.07025	0.07198	0.07716	0.08723	0.13014

§5. Summary and discussion

In order to have an adiabatic change of the magnetization when the strength of the magnetic field is changed continuously in time, it is necessary for there to exist avoided level crossings. An avoided level crossing is caused by some interaction that does not commute with the magnetization, such as the Dzyaloshinskii-Moriya interaction (DMI). However, the effect of the DMI depends on the direction of the magnetic field, and for certain directions of the applied field, the DMI does not generate avoided level crossing structure.

As an alternative source of avoided level crossings, we have studied energy gap formation due to the hyperfine interaction. For this mechanism, the gap is independent of the direction of the applied field.

We found that adiabatic transitions that start from the ground state can be analyzed using the Landau-Zener-Stückelberg (LZS) formula with an effective energy gap. We studied the change of magnetization at finite temperature, assuming a canonical distribution of the initial level occupation. The degree to which the change was adiabatic was estimated from the change of the magnetization. We found that this depends weakly on the temperature, as long as the temperature is of the same order as the strength of the hyperfine interaction. Also, the dependence on the rate of change of the field strength can be fitted using the LZS formula with an effective energy gap.

Our results suggest that apparent LSZ transitions can occur even if the energy level structure does not consist of pairs of repelling levels. Therefore it is necessary to study whether the observed experimental results are due to a true LZS mechanism of a well-defined energy structure or an apparent LSZ mechanism. Usually, the gap due to the hyperfine interaction is believed to be much smaller than that due

to the electron spin interaction. However, for an energy gap of order 10^{-2}K, the hyperfine interaction could play a significant role in opening the gap. With this in mind, we have to reconsider the source of the adiabatic changes of magnetization, i.e., temperature independent, smooth changes of the magnetization caused by a continuously changing field strength. Actually, it has been pointed out that a simple DM interaction cannot account for the adiabatic transitions in V_{15} [6] and Fe_6. [14] The present study may help to resolve this problem.

Describing the coherence of the state in this apparent LSZ transition is an interesting problem on which we will report elsewhere. Clarifying the effect of contact with a thermal bath, in particular, the manner in which the magnetic Föhn effect appears in this system, is another interesting problem, which is also left for future research.

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