Kinetics of decay of metastable polarized crystals

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We develop a theory that describes the principal channels of the decay of metastable polarized crystals–crystals made up of atoms with parallel electron spins. It is shown that the decay of these crystals is connected principally with the production of unique spin waves (magnons) that carry negative energy. The principal decay channels, however, can be suppressed by using an external magnetic field directed opposite to the polarization of the electron magnetic moments. The leading decay channels, which cannot be eliminated by the magnetic field, turn out to be connected with the decay of the polarized state to one magnon and two phonons, on account of the dipole–dipole interaction of the electron spins, and into one magnon and one phonon on account of a second-order process due to simultaneous hyperfine interaction and magnon–phonon interaction of exchange character. In the case of polarized alkali-atoms crystals the first type of interaction leads to a decay probability per particle $\sim 10^{-3}$–10^{-4} sec^{-1}, and determines by the same token the rather long lifetime. The second channel turns out to be relatively faster with a characteristic decay probability, $\sim 10^{-1}$–10^{-3} sec^{-1}. It can, however, be suppressed by simultaneous polarization of the nuclear spins.

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1. INTRODUCTION

The problem of metastable states of matter, which result from polarization of the electron spins of the atoms in the system, has been attracting great interest of late. If the interaction between the atoms depends substantially on the exchange, then the spin configuration that appears upon polarization can lead to formation of a metastable phase. Of greatest interest, particularly from the point of physical clarity, are the atomic phases of hydrogen and alkali metals. A preliminary analysis[1] has shown[1)] that at low temperatures polarized atoms of alkali metals condense into a metastable crystalline phase that is locally stable at a pressure $P = 0$. The attraction between the atoms in this phase is due to van der Waals forces, and this, just as in the case of solidifying gases, leads to dielectric crystals with large interatomic distances and relatively small sublimation energy.

A similar situation obtains in the case of atomic hydrogen, the only difference being that in this case it may be necessary to have a finite pressure, to form a metastable polarized crystal.

The decisive circumstance for the attaining and using such states is, naturally, the question of their lifetimes. We immediately encounter here the principal difference between the systems considered here and ordinary metastable states. In the standard cases the stable and metastable phases are separated by a barrier in configuration space, and the metastable phase can decay only via formation of germs of critical dimension. In the metastable polarized crystals the kinetics of the decay is connected with spin flip of individual atoms or with the appearance of single collective spin excitations. There is no surface energy here, and a large lifetime can be reached only on account of a low probability of spin flip of the atoms. For this, however, there are special grounds.

When the spin of an individual atom flips, the potential relief in which this atom is situated changes rapidly. Instead of being in a potential well, the atom turns out to be on a potential hump. If the difference between the interaction energy at total polarization (the bottom of the well) and in the presence of one flipped spin (the peak of the hump) is large enough compared with the Debye energy of the crystal, then the amplitude of the transition is exponentially small. Simultaneously with this, however, an important role is played by inelastic processes with phonon emission; these processes are due to the jarring experienced by the crystal lattice at the instant of the spin flip. This leads to a noticable increase of the transition probability. The picture that arises in the case of local spin flip and the calculation of the summary transition probability in this case are the subject of the third section of the present paper. As will be seen from this analysis, at realistic values of the parameters of the problem the probability of the transition can be quite small, and then this decay channel offers no danger. At the same time, the answer is most sensitive to the values of the parameters of the problem, by virtue of which the corresponding lifetime can vary greatly from substance to substance.

The metastable crystal in question constitutes a pseudoferromagnetic system described by a Heisenberg Hamiltonian. In such a system, the spin flip can be realized via production of collective excitations—magnons. The peculiarity in this case consists in the fact that the magnons will have negative energy. The next problem is therefore the analysis of the decay of the metastable polarized crystal via production of collective excitations.

It must be stated that the real danger is posed by the production of relatively long-wave magnons, particularly together with phonons. A detailed analysis of the decay of the crystal in this case and a determination of the corresponding lifetimes, are given in Secs. 4 and 5, where arguments that demonstrate the possibility of closing the most dangerous decay channels are also developed. It should be noted in this connection that a recently published note[2] contains the statement that a

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crystal made up of polarized hydrogen atoms has a "fast" decay channel connected with production of one or two magnons. It will be shown below that this decay channel can be suppressed.

2. INITIAL PREMISES

Consider a crystal considering of univalent atoms with the electron in the S state. The interaction of an isolated pair of such particles at distances R large compared with the atomic distance a_0 can be expressed in the form

$$\hat{v}(R)=U(R)+\Delta(R)\left({}^1/_4+\hat{\mathbf{S}}_1\hat{\mathbf{S}}_2\right). \tag{2.1}$$

Here $U(R)$ is the long-range multipole interaction between the atoms and corresponds to their attraction. The second term in (2.1) describes the exchange interaction due to overlap of the electron shells of the atom. $\Delta(R)$ is equal to the difference between the energies of the triplet and the singlet states of the considered pair of atoms, and $\hat{\mathbf{S}}_1$ and $\hat{\mathbf{S}}_2$ are the electron-spin operators.

The main contribution to $\Delta(R)$ is made by the electron density localized near the axis joining the nuclei.[3,4] Consequently, if the interatomic distance in the crystal is large compared with a_0, then the multiparticle interaction between atoms in the crystal lattice can be represented as a sum of pair interactions of the form (2.1):

$$\hat{V}=\frac{1}{2}\sum_{l,m}U(R_{lm})+\frac{1}{2}\sum_{l,m}\Delta(R_{lm})\left(\frac{1}{4}+\hat{\mathbf{S}}_l\hat{\mathbf{S}}_m\right). \tag{2.2}$$

In a system of atoms with parallel spins, the second term corresponds to repulsion. The minimum of the potential energy (2.2) is realized as a rule in a lattice of the fcc or hcp type. In all the cases of interest to us, at distances a between the nearest atoms, the following inequality turns out to be realistic

$$a\gg a_0. \tag{2.3}$$

Thus, for a crystal made up of polarized Na atoms a direct calculation yields $a=4.9$ Å, and accordingly the sublimation energy per atom is $\varepsilon=2000$ K. Values of the same order are obtained also for crystals of other alkali atoms (all the calculations that follow are based on the Na crystal parameters obtained by Shlyapnikov and Shmatov).

The calculation of the phonon spectrum, starting from the interaction (2.2), yields in all cases for the mean squared displacement of the atom the relation

$$\overline{u^2}/a^2|\ll 1, \tag{2.4}$$

so that the crystal remains classical and we can confine ourselves in the estimates that follow to the harmonic approximation.

3. LOCAL SPIN FLIP

In the considered metastable polarized crystals there exist two types of interaction that can lead to the flip of the electron spin of an individual atom. The first is the hyperfine interaction with the spin of its own nucleus, and the other is dipole–dipole interaction with electron spins of neighboring atoms. The large value of the interatomic distance leads in general to a situation wherein both interactions are comparable in magnitude.

We assume first that the surrounding atoms are rigidly fixed. Then, according to (2.2), the atom with the flipped spin turns out to be in a potential relief determined only by the multipole part of the interaction, while the potential energy changes by an amount

$$\delta V=-\frac{1}{2}\sum_{g}\Delta(\mathbf{R}_g^0-\mathbf{R}), \tag{3.1}$$

if we confine ourselves to interaction with only the nearest neighbors (index g). The vector $\mathbf{R}$ characterizes the position of the atom whose spin was flipped, and is reckoned from the equilibrium position $R=0$. The energy gap between the bottom of the potential well in which the atom is located prior to the spin flip and the peak of the resultant potential hump at $R=0$ (see the figure) is

$$E_0=Z\Delta_0/2,\quad \Delta_0=\Delta(a), \tag{3.2}$$

where Z is the number of nearest neighbors.

To estimate the matrix element of the transition, we choose the wave function of the initial state in the usual oscillator form:

$$\Psi_1(R)=\frac{2}{\pi}\left(\frac{M\omega_0\pi}{\hbar}\right)^{3/4}\times\exp\left(-\frac{M\omega_0}{2\hbar}R^2\right) \tag{3.3}$$

which is the wave function of the ground state of a three-dimensional oscillator (M is the mass of the atom).

Using crystals of alkali-metal atoms as an example, we can easily ascertain that always

$$E_0\gg\hbar\omega_1,\ \hbar\omega_0, \tag{3.4}$$

where ω_1 is determined in terms of the modulus of the second derivative of the potential hump. In fact we always have

$$\omega_1\ll\omega_0. \tag{3.5}$$

Therefore the motion of the particle with the flipped spin near $R=0$ has a quasiclassical character and the corresponding wave function for the S state can be written in the form

$$\Psi_2(R)=\left[\frac{2M}{\pi\hbar g(E)P_2(R)}\right]^{1/2}\frac{1}{R}\sin\left(\frac{1}{\hbar}\int_0^R P_2(R')\,dR'\right) \tag{3.6}$$

(at small R we assume the potential to be spherically symmetrical). Here $g(E)=dn/dE$ is the quasiclassical state density, E is the particle energy in the final state and is reckoned from the peak of the potential hump, and $P_2(R)$ is its classical momentum. The wave functions (3.3) and (3.6) are normalized by the condition

$$\int_0^\infty|\Psi_{1,2}(R)|^2R^2\,dR=1.$$

The matrix element of the transition with local spin flip contains as a factor the overlap integral

$$J(E)=\int_0^\infty\Psi_1(R)\Psi_2(R)R^2\,dR. \tag{3.7}$$

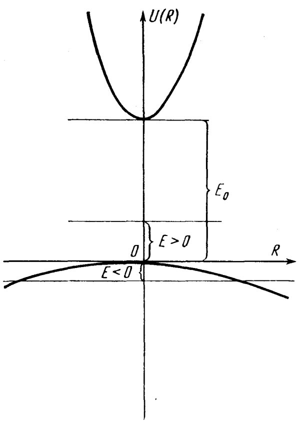

FIG. 1. The potential energy of the interaction of an individual crystal atom with its surroundings (R is the displacement of the atom from its equilibrium position). The potential well corresponds to the state of total polarization of the electron spins, while the potential hump corresponds to a state in which the spin of the given atom is flipped.

Substituting here the expressions (3.3) and (3.6), we get

$$J(E)=2\left(\frac{2E}{\pi}\right)^{1/4}\exp\left(-\frac{E}{\hbar\omega_0}\right)\Big/(\hbar\omega_0)^{3/4}(g(E))^{1/2}. \tag{3.8}$$

We denote the matrix element of the interaction that leads to the local flip of the electron spin by $(H_{\text{int}})_{12}$. We then have for the probability of such a transition, after integrating over the final states of the atom with the flipped spin,

$$W=16\omega_0\left|\frac{(H_{int})_{12}}{\hbar\omega_0}\right|^2\left(\frac{\pi E_0}{2\hbar\omega_0}\right)^{1/2}\exp\left\{-\frac{2E_0}{\hbar\omega_0}\right\}. \tag{3.9}$$

In the case of the hyperfine interaction, an estimate yields (10^6-10^9) sec^{-1} for the pre-exponential factor in (3.9) for alkali-atom crystals. In a rigid lattice, a dipole-dipole interaction, owing to symmetry, does not lead at all to local spin flip of an individual atom. The matrix element $(H_{\text{int}})_{12}$ turns out to be different from zero only when account is taken of the displacement of the atoms, and this leads to an additional small factor (2.6), so that the pre-exponential factor in (3.9) does not exceed in this case the values given above.

In all cases, with the exception of atomic hydrogen, which is not considered in the present paper, the argument of the exponential in (3.9) is very large

$$\Delta_0/\hbar\omega_0\gg 1. \tag{3.10}$$

Thus, for a sodium-atom crystal $\Delta_0/\hbar\omega_0=12$. Values of the same order are obtained also for crystals of other alkali atoms. The transition probability in this case is so small, that in the absence of excitation of the phonon subsystem the polarized crystal turns out to be practically stable to spin flip of an individual atom.

We now dispense with the assumption that the lattice is rigid and consider the jarring effect, which inevitably takes place when the potential energy changes by δV (3.1). This additional interaction leads to a shift of the positions of the phonon-field oscillators. Neglecting the renormalization of the phonon spectrum and retaining the representation corresponding to the initial undistorted lattice of the crystal, the allowance for such a force impact in the lattice, as is well known, is equivalent to introducing the shift operator

$$\hat{T}=\exp\left\{\sum_\beta\frac{A_\beta}{\hbar\omega_\beta}(\hat{b}_\beta+\hat{b}_\beta{}^+)\right\}. \tag{3.11}$$

Here $\hat{b}_\beta$ and $\hat{b}_\beta^+$ are the operators for the annihilation and production of a phonon with index $\beta\equiv\mathbf{q}$, α characterizing the value of the wave vector $\mathbf{q}$ and the branch number α, while ω_β is the phonon frequency. The coefficients A_β are obtained directly from the linear expansion of (3.1) and the displacements of the nearest atoms. Taking symmetry into account we have

$$A_\beta=-\frac{i}{2}\sum_g\left[\frac{\hbar}{2M\omega_\beta N}\right]^{1/2}\Delta_0'\frac{\mathbf{g}\mathbf{e}_\beta}{a}\sin(\mathbf{q}\mathbf{g}), \tag{3.12}$$

$$\Delta_0'=d\Delta(a)/da,$$

where $\mathbf{g}\equiv\mathbf{R}_g^0$, $\mathbf{e}_\beta$ is the phonon polarization vector, and N is the number of crystal atoms. The matrix element, diagonal in the phonons, of the operator (3.11) is equal to

$$T_0=\exp\left\{-\frac{1}{2}\sum_\beta\left(\frac{A_\beta}{\hbar\omega_\beta}\right)^2(2\bar{n}_\beta+1)\right\}=\exp\left[-\frac{1}{2}\Phi(T)\right], \tag{3.13}$$

where $\bar{n}_\beta$ is the average temperature value of the occupation numbers and T is the crystal temperature.

In the Debye approximation for the phonon spectrum we have

$$\Phi_0=\Phi(T=0)\approx\frac{3Z}{4}\left(\frac{\Delta_0}{\hbar\omega_D}\right)^2\frac{\hbar}{M\omega_D}\left(\frac{\Delta_0'}{\Delta_0}\right)^2. \tag{3.14}$$

The probability of a local spin flip elastic in the phonons reduces in this case to expression (3.9) multiplied by T_0^2. For metastable polarized crystals of alkali atoms we have

$$\Phi_0\gg 1, \tag{3.15}$$

and this probability acquires an additional smallness.

Now, however, the inelastic processes become important. For the probability of spin flip accompanied by transfer of an energy E_0-E to the phonon subsystem, using the known transformations, we have

$$W(E)=\frac{2\pi}{\hbar}|(H_{int})_{12}|^2J^2(E)\sum_{\nu,\mu}\rho_{ph}^{(0)}(\nu)|T_{\nu\mu}|^2$$
$$\times\delta(E_\nu-E_\mu+E_0-E)=\left|\frac{(H_{int})_{12}}{\hbar}\right|^2J^2(E)e^{-\Phi}$$
$$\times\int_{-\infty}^{+\infty}dt\exp\left\{\chi(t)+\frac{i(E_0-E)}{\hbar}t\right\}, \tag{3.16}$$

where

$$\chi(t)=\sum_\beta\left(\frac{A_\beta}{\hbar\omega_\beta}\right)^2[(\bar{n}_\beta+1)e^{-i\omega t}+\bar{n}_\beta e^{i\omega t}], \tag{3.17}$$

$\rho_{\text{ph}}^{(0)}(\nu)$ is the equilibrium phonon density matrix, E_ν is the energy of the phonon subsystem in the state ν. The energy E is reckoned from the peak of the potential hump (see the figure).

We confine ourselves to the case of sufficiently low temperatures when, in particular, $\Phi\approx\Phi_0$. When (3.15) is taken into account, the minimum of the integral in

(3.16) occurs in the region $E_0 - E \sim \hbar\omega_D\Phi_0$. Therefore the effective values of t in the integral correspond to the condition $\omega_D t \ll 1$, and we can use the expansion of $\chi(t)$ at small t:

$$\chi(t) = \Phi_0 - it\Phi_0\gamma_1\omega_D - {}^1/_2 t^2\Phi_0\gamma_2\omega_D^2. \tag{3.18}$$

We have introduced here the notation

$$\sum_\beta \left(\frac{A_\beta}{\hbar\omega_\beta}\right)^2 (\omega_\beta)^k = \Phi_0\gamma_k(\omega_D)^k. \tag{3.19}$$

Substituting (3.18) in (3.16), we get

$$\begin{gathered}\int_{-\infty}^{+\infty} dt\exp\left\{\chi(t) + \frac{i(E_0-E)}{\hbar}t\right\} = \left(\frac{2\pi}{\Phi_0\omega_D^2\gamma_2}\right)^{1/2}\exp(\Phi_0)\\ \times\exp\left\{-\left(\frac{E_0}{\hbar\omega_D} - \Phi_0\gamma_1 - \frac{E}{\hbar\omega_D}\right)^2\Big/2\Phi_0\gamma_2\right\}.\end{gathered} \tag{3.20}$$

We now determine the total probability of the inelastic transition, at which the particle remains in states above the barrier:

$$W_1 = \int_0^{E_0} dE\, g(E)W(E). \tag{3.21}$$

We substitute in (3.16) the expression (3.8) and consider first the most characteristic case, when $E_0 > \Phi_0\hbar\omega_D$. Taking (3.15) into consideration and using the saddle-point method, we obtain

$$\begin{gathered}W_1 = 8\pi\omega_D\left|\frac{(H_{int})_{12}}{\hbar\omega_D}\right|^2\left(\frac{2x^*}{\pi}\right)^{1/2}\left(\frac{\omega_D}{\omega_0}\right)^{3/2}\\ \times\exp\left\{-\frac{2E_0}{\hbar\omega_0} + 2\Phi_0\left[\gamma_1\frac{\omega_D}{\omega_0} + \gamma_2\left(\frac{\omega_D}{\omega_0}\right)^2\right]\right\}.\end{gathered} \tag{3.22}$$

Here

$$x^* = \frac{E_0}{\hbar\omega_D} - 2\Phi_0\gamma_2\frac{\omega_D}{\omega_0} - \Phi_0\gamma_1 \tag{3.23}$$

is the position of the extremum of the argument of the exponential in the integrand of (3.21). The result is valid under the condition

$$(x^*)^2 \gg 2\Phi_0\gamma_2. \tag{3.24}$$

In the Debye model we have

$$\gamma_1 \approx 0.65, \quad \gamma_2 \approx 0.35, \quad \omega_D/\omega_0 = ({}^5/_3)^{1/2}.$$

With increasing Φ_0, the probability of spinflip increases continuously because of the inelastic processes accompanying the jarring. However, if Φ_0 remains smaller than $E_0/\hbar\omega_D$, then under the condition (3.24) the crystal lifetime, defined as the reciprocal of (3.22), turns out to be very large. For a crystal made up of Na atoms we have $\Phi_0 \approx 15$ and the condition (3.24) is satisfied quite well. The argument of the exponential in (3.22) is $\sim 10^2$, i.e., the crystal is practically stable at low temperature relative to a local spin flip. A similar situation obtains apparently also for metastable polarized crystals of other alkali atoms.

In the general case, when Φ_0 increases the value of x^* (3.23) shifts towards the lower limit in the integral of (3.21) and then reverses sign. With further increase of Φ_0, the main contribution to this integral is due only to the region near the lower limit. Direct calculations at

$$|x^*| \gg (\Phi_0\gamma_2)^{1/2} \tag{3.25}$$

yield

$$W_1 = 4\omega_D\left(\frac{\pi\omega_D}{|x^*|\omega_0}\right)^{3/2}\frac{\Phi_0\gamma_2}{\pi}\left|\frac{(H_{int})_{12}}{\hbar\omega_D}\right|^2\exp\left\{-\frac{(E_0/\hbar\omega_D - \Phi_0\gamma_1)^2}{2\Phi_0\gamma_2}\right\}. \tag{3.26}$$

In the considered region of the values of the parameters, when $\hbar\omega_D\Phi_0 > E_0$, transitions of a particle in states lying below the peak of the potential hump can become substantial. In this case by virtue of (3.25), the value of the overlap integral (3.7) is connected with the behavior of the wave function $\Psi_2(R)$ in the region below the barrier. Determining $\Psi_2(R)$ in the quasiclassical approximation for a parabolic potential hump, we easily obtain by taking (3.3) and (3.5) into account

$$J^2(E) \sim \frac{|E|^{1/2}}{g(E)}\exp\left(-\frac{\pi|E|}{\hbar\omega_1}\right), \quad E<0.$$

Substituting this expression in (3.16) and calculating the total transition probability for the region $E<0$

$$W_2 = \int_{-\infty}^{0} dE\, g(E)W(E),$$

we can easily verify that the main contribution to this integral is made by an energy interval near the upper limit, thereby predetermining the appearance of the same exponential as in (3.26), but with a substantially pre-exponential factor. As a result, the total transition probability W is determined in practice by the value of W_1 (3.26).

If $\hbar\omega_D\Phi_0 > E_0$ and the condition (3.25) is satisfied, then the argument of the exponential in (3.26) is large and the lifetime of the metastable polarized crystal relative to a large spin flip of an individual atom can be very large. Thus, in both limiting cases the crystal turns out to be sufficiently stable to the considered decay channel. At the same time if $E_0/\hbar\omega_D$ is close enough to Φ_0 [see (3.22) and (3.25)], this decay channel can present a substantial danger.

4. SPIN WAVES IN METASTABLE POLARIZED CRYSTALS

A small probability of local spin flip still does not mean that the metastable polarized crystal is long-lived. There can appear in such a crystal nonlocalized spin excitations—peculiar spin waves (magnons) that carry negative energy. The last circumstance means that the magnon production is a transition of the crystal to a lower energy state. The energy released thereby goes to phonon excitation. The greatest danger is presented by the production of long-wave magnons. This is due to two causes. On the one hand, there is no need for the production of a large number of phonons, which would lead to a sharp decrease of the transition probability. On the other hand, the long-wave magnons interact relatively weakly with the phonons, and this leads to a weak damping of these magnons and eliminates the problem of their formation. It must be stated that for the parameter ratio assumed in this paper we can make a much stronger statement, namely, that weak damping does in fact take place for magnons of arbitrary wavelength. To a decisive degree this is due to the inequality (3.10).

In the second-quantization representation the Hamiltonian (2.2) for the magnons can be written in standard form

$$\hat{H}=\sum_{\mathbf{f}}\varepsilon_{\mathbf{f}}\hat{a}_{\mathbf{f}}^{+}\hat{a}_{\mathbf{f}}+\text{const.} \tag{4.1}$$

Here $\hat{a}_{\mathbf{f}}^{+}$ and $\hat{a}_{\mathbf{f}}$ are the operators for the creation and annihilation of a magnon with wave vector f, whose energy is

$$\varepsilon_{\mathbf{f}}=-\frac{1}{2}\sum_{\mathbf{g}}\Delta_0(1-\cos\mathbf{fg}). \tag{4.2}$$

The Hamiltonian of the interaction of the spin waves with the phonons is obtained directly from (2.2) if the exchange interaction Δ is expanded in the relative displacements of the atoms. Confining ourselves to the linear term of the expansion, we have

$$\hat{H}_1=\sum_{\beta,\mathbf{f}}A_{\beta\mathbf{f}}\hat{a}_{\mathbf{f}-\mathbf{q}}^{+}\hat{a}_{\mathbf{f}}[\hat{b}_{\beta}+\hat{b}_{-\beta}^{+}]. \tag{4.3}$$

$$A_{\beta\mathbf{f}}=-\frac{\Delta_0'}{2}\left(\frac{\hbar}{2M\omega_{\beta}N}\right)^{1/2}\sum_{\mathbf{g}}\frac{\mathbf{e}_{\beta}\mathbf{g}}{a}(1-e^{-i\mathbf{qg}})(1-e^{i\mathbf{fg}}), \tag{4.4}$$

where Δ_0' is defined in accordance with (3.12).

As a result of the interaction (4.3) the spin wave is accompanied by a change in the state of the lattice. The scale of the corresponding jarring effect can be estimated in this case by finding the overlap integral between the crystal vibrational wave functions Ψ_0 and $\Psi_{0\mathbf{f}}$ corresponding respectively to total polarization and to the presence of one magnon with conservation of the phonon occupation numbers.

We determine the function $\Psi_{0\mathbf{f}}$ in second-order perturbation theory in the Hamiltonian $\hat{H}_1$ (4.3). Then ($T\approx 0$)

$$J_{\mathbf{f}}=\langle\Psi_0|\Psi_{0\mathbf{f}}\rangle=1-\frac{1}{2}\sum_{\beta}\frac{|A_{\beta\mathbf{f}}|^2}{(\varepsilon_{\mathbf{f}-\mathbf{q}}-\varepsilon_{\mathbf{f}}+\hbar\omega_{\beta})^2}\equiv 1-\frac{1}{2}G_{\mathbf{f}}. \tag{4.5}$$

Comparing $G_{\mathbf{f}}$ with expressions (3.13) and (3.14) and taking relations (4.4) and (3.12) into account, we can easily obtain the following estimate for a magnon with arbitrary f:

$$G_{\mathbf{f}}\sim 4\Phi_0\left(\frac{4\hbar\omega_D}{Z\Delta_0}\right)^2\frac{|\varepsilon_{\mathbf{f}}|}{Z\Delta_0}. \tag{4.6}$$

As we have seen in the preceding section, $\Phi_0 \gg 1$. For crystals of alkali-metal atoms, however, the ratio $\hbar\omega_D/\Delta_0$ turns out to be so small that it cancels out the value Φ_0 and we have in (4.6)

$$G_{\mathbf{f}}\ll 1$$

for spin waves with arbitrary f. Thus, for the Na crystal we have $G_f \sim 10^{-2}|\varepsilon_f|/\Delta_0$. In the case of long-wave magnons ($fa \ll 1$) we have

$$\varepsilon_{\mathbf{f}}=-\frac{Z\Delta_0}{12}(fa)^2 \tag{4.7}$$

and the parameter G_f acquires an additional smallness.

Actually Δ_0 and $\hbar\omega_D$ (and consequently also Φ_0) are not independent parameters. Thus, $\omega_D^3 \sim Z\Delta_0''/M$. Recognizing that $\Delta_0'^2/\Delta_0\Delta_0'' \sim 1$, we obtain the estimate

$$\Phi_0\sim 3\Delta_0/\hbar\omega_D. \tag{4.8}$$

Consequently, at an inverse ratio of the parameters ($\hbar\omega_D \gg Z\Delta_0$) the quantity Φ_0 becomes less than 1. At the same time, in this limit it follows from (4.5) and (3.14) that

$$G_{\mathbf{f}}\sim 4\Phi_0|\varepsilon_{\mathbf{f}}|/Z\Delta_0$$

and again $G_f \ll 1$ for magnons of arbitrary wavelength. In a noticeable parameter interval, however, when $\hbar\omega_D \sim Z\Delta_0$, the value of G_f can be of the order of unity. Nonetheless, even in this case $G_f \ll 1$ at $fa \ll 1$, so that for long-wave magnons the "polaron" effect is weak and effects of excitation of the phonon subsystem can be taken into account by perturbation theory.

We note that for the same reasons the magnons are long-lived collective excitations. A direct estimate of their lifetime yields

$$\frac{1}{\tau_{\mathbf{f}}}\sim\frac{Z\Delta_0}{4\hbar}G_{\mathbf{f}}. \tag{4.9}$$

For long-wave magnons there is added to the right-hand side of (4.9) a small factor $\min\{(fa)^4, (\hbar\omega_D/\Delta_0)^4\}$.

5. DECAY OF CRYSTAL VIA EXCITATION OF SPIN WAVES

A. Single-magnon and two-magnon excitation

Production of spin waves in a metastable polarized crystal is due to two types of interaction: hyperfine interaction

$$\begin{aligned}\hat{H}'&=\frac{A}{2}\sum_{m}(\hat{\mathbf{S}}_m^{+}\hat{\mathbf{I}}_m^{-}+\hat{\mathbf{S}}_m^{-}\hat{\mathbf{I}}_m^{+})\\&=\frac{A}{2N^{1/2}}\sum_{m,f}(\hat{a}_{\mathbf{f}}^{+}\hat{\mathbf{I}}_m^{-}e^{i\mathbf{fm}}+\hat{a}_{\mathbf{f}}\hat{\mathbf{I}}_m^{+}e^{-i\mathbf{fm}})\end{aligned} \tag{5.1}$$

(I is the nuclear spin operator, ($\mathbf{m}\equiv\mathbf{R}_{m0}$), and dipole–dipole interaction of the electron spins

$$\hat{H}''=2\mu_B^2\sum_{m,l}\frac{(\hat{\mathbf{S}}_m\hat{\mathbf{S}}_l)R_{ml}^2-3(\hat{\mathbf{S}}_m\mathbf{R}_{ml})(\hat{\mathbf{S}}_l\mathbf{R}_{ml})}{R_{ml}^5}. \tag{5.2}$$

We note that in the crystals under consideration $a/a_B \sim 10$, and consequently the interactions (5.1) and (5.2) turn out to be comparable in magnitude. The interaction (5.2) contains the terms responsible for the production of one and two magnons, and also terms that do not change the number of magnons. The latter, however, are small compared with the corresponding terms in the Hamiltonian $\hat{H}_1$ (4.3).

Transitions with excitation of one magnon can be produced by either interaction (5.1) or (5.2). In the absence of phonon excitation, the energy conservation law can be satisfied in the presence of an external magnetic field. If the magnetic moments of the electrons are polarized along the field, then

$$\varepsilon_{\mathbf{f}}=2\mu_B H-{}^1/_{12}Z\Delta_0(fa)^2. \tag{5.3}$$

The dipole–dipole interaction (5.2) does not lead to production of one magnon without simultaneous excitation of phonons, by virtue of the momentum conservation law. In the case of the hyperfine interaction (5.1), the local spin flip of the nucleus eliminates the momentum-conservation problem and leads to production of one magnon with a wave vector determined from the condition $\varepsilon_f = 0$. In first-order perturbation theory we have for the probability of one-magnon decay per particle

$$W = \frac{12I(I+1)}{\pi} \left(\frac{\Omega_0}{a^3}\right) \left(\frac{Z\mu_B H}{6\Delta_0}\right)^{1/2} \left(\frac{A}{Z\Delta_0}\right)^2 \frac{\Delta_0}{\hbar} \tag{5.4}$$

[averaging over the projection of the nuclear spin is carried out in (5.4), and Ω_0 is the volume of the unit cell].

The dipole-dipole interaction leads to a decay accompanied by the production of two magnons with oppositely directed momenta. For the probability of this process, a direct calculation yields

$$W = \frac{6\pi}{5} \left(\frac{a^3}{\Omega_0}\right) \left(\frac{Z\mu_B H}{6\Delta_0}\right)^{1/2} \left(\frac{8\mu_B^2}{Za^3\Delta_0}\right)^2 \frac{\Delta_0}{\hbar}. \tag{5.5}$$

The presence of analogous decay channels in a metastable polarized crystal of atomic hydrogen was first pointed out by Berlinsky *et al.*[2] In the case of crystals of alkali atoms, formulas (5.4) and (5.5) yield close estimates

$$W \sim (10^3 - 10^5) \left(\frac{\mu_B H}{\Delta_0}\right)^{1/2} \ [\mathrm{sec}^{-1}]. \tag{5.6}$$

Although the probabilities (5.4)–(5.6) contain a small parameter $\mu_B H/\Delta_0$, they correspond to channels of a relatively fast decay, for in the case of polarization along the magnetic field H can realistically not be chosen to be lower than the intrinsic field of the sample. The corresponding lifetimes turn out to be in this case $\tau \sim 10^{-1}$–10^{-3} sec.

However, these channels will be in general forbidden if there is a sufficiently external magnetic field directed opposite to the polarization of the magnetic moments. An experimental realization of such a situation entails no difficulties in principle. In particular, it is usually realized in a film of polarized atomic hydrogen produced by deposition of an atomic beam on a cold substrate.

B. Decay via excitation of magnons and phonons

The problem of energy conservation is eliminated to a considerable degree if phonons are emitted simultaneously with the magnons. We consider transitions of this kind, due to dipole-dipole interaction. The simplest mechanism is the simultaneous production of one magnon and one phonon. The linear term of the expansion of the Hamiltonian (5.2) in the atom displacements, which corresponds to this decay channel, can be easily reduced to the form

$$\hat{H}_1'' = -2i \sum_{\beta,\mathbf{l}} \left(\frac{\hbar}{2M\omega_\beta}\right)^{1/2} \sin \mathbf{q}\mathbf{l} \frac{dV^d(\mathbf{R}_l)}{d\mathbf{R}_l} \mathbf{e}_\beta \hat{a}_\mathbf{q}^+ \hat{b}_\beta^+, \tag{5.7}$$

where

$$V^d(\mathbf{R}_l) = \frac{3\mu_B^2}{2} \frac{z_l(x_l - iy_l)}{R_l^5}$$

(z is the axis along which the electron magnetic moments are directed).

The momentum conservation law $\mathbf{f} = \mathbf{q}$ [it was used to obtain the explicit form of (5.7)] and the energy conservation law result, under the condition (3.10) in the production of only a long-wave magnon and a long-wave phonon in the process under consideration. Taking this into consideration, we obtain in first-order perturbation theory for the decay probability

$$W \approx \left(\frac{3\mu_B^2}{2a^3\Delta_0}\right)^2 \left(\frac{\hbar\omega_D}{Z\Delta_0}\right)^2 \left(\frac{\hbar}{M\omega_D a^2}\right) \frac{Z\Delta_0}{\hbar}. \tag{5.8}$$

An estimate of this expression for alkali-atom crystals yields $\tau \sim 10^2$–10^4 sec.

We note that the probability of simultaneous production of two magnons and one phonon on account of dipole–dipole interaction turns out to be much less than the probability (5.8). This process arises also in second-order perturbation theory when the Hamiltonians (5.2) and (4.3) are used. Although in this case the probability of the transition is larger than when account is taken of only the dipole–dipole interaction, for the crystals considered by us it is always less than the probability (5.8).

In principle, any single-phonon decay channel can also be suppressed by a sufficiently strong external magnetic field directed against the polarization of the magnetic moments. Thus, in the case of a process with simultaneous production of one magnon we can usually obtain from the energy conservation law

$$\tfrac{1}{12} Z\Delta_0 (qa)^2 + 2\mu_B H = \hbar\omega_{\mathbf{q}\alpha}$$

an estimate for the corresponding value of the field

$$H > H_{\mathrm{cr}} \sim \frac{\Delta_0}{\mu_B} \left(\frac{\hbar\omega_D}{Z\Delta_0}\right)^2.$$

In crystals made up of alkali-metal atoms, the critical field turns out to be relatively weak:

$$H_{\mathrm{cr}} \sim 10^2 - 10^3 \ \mathrm{Oe}.$$

The possibility of suppressing the decay channels caused by the dipole–dipole interaction is limited to the case of single-phonon excitations. If a larger number of phonons is involved in the decay process, then the conservation laws can be satisfied in fact at an arbitrary value of H. The greatest danger here, naturally, lies in the decay channel corresponding to production of one magnon and two phonons. The interaction Hamiltonian corresponding to this process is obtained by taking into account the terms quadratic in the atom displacements in the expansion (5.2):

$$\hat{H}_2'' = \sum_{\beta_1,\beta_2,\mathbf{l}} \frac{\hbar}{2M(\omega_{\beta_1}\omega_{\beta_2})^{1/2}} (1 + \cos(\mathbf{q}_1 + \mathbf{q}_2)\mathbf{l} - \cos \mathbf{q}_1\mathbf{l} - \cos \mathbf{q}_2\mathbf{l}) \times \frac{d^2V^d(\mathbf{R}_l)}{dR_l^i\, dR_l^k} e_{\beta_1}{}^i e_{\beta_2}{}^k \hat{a}^+_{\mathbf{q}_1+\mathbf{q}_2} \hat{b}_{\beta_1}{}^+ \hat{b}_{\beta_2}{}^+. \tag{5.9}$$

In this expression the momentum conservation law is taken into account in the form

$$\mathbf{f} = \mathbf{q}_1 + \mathbf{q}_2.$$

The energy conservation law again leads to excitation of only long-wave magnons. Now, however, there are no substantial limitations on the phonon energy, and the principal role is assumed by phonons with large momenta, and $\mathbf{q}_1+\mathbf{q}_2\approx 0$. Using in the calculations the Debye model of the phonon spectrum, we obtain for the transition probability

$$W\approx\left(\frac{3\mu_B{}^2}{4a^3\Delta_0}\right)^2\left(\frac{\hbar\omega_D}{Z\Delta_0}\right)^{1/2}\left(\frac{\hbar}{M\omega_D a^2}\right)^2\frac{Z\Delta_0}{\hbar}. \qquad (5.10)$$

In the case of alkali-metal crystals an estimate of this expression yields

$$\tau\sim 10^3-10^4 \text{ sec}. \qquad (5.11)$$

The involvement of a large number of phonons in the decay process leads to the appearance of a small factor $\overline{u}^2/a^2$ for each additional phonon. Therefore the characteristic lifetime is precisely the one determined by formula (5.10) which turns out, as follows from (5.11), to be quite large.

Attention should be called to the fact that two-phonon processes appear when account is taken of higher orders of perturbation theory, when the virtual production of two magnons or of a magnon and a phonon on account of the dipole–dipole interaction is accompanied by a rescattering of the magnons and by the emission of a phonon as a result of the interaction (4.3), which is of exchange origin. Direct calculations of the probabilities of such processes show that in the case of crystals of alkali-metal atoms it is lower than the probability (5.10).

The hyperfine interaction (5.1) does not depend on the displacements of the atoms and in itself does not lead to phonon excitation. It can, however, lead to the production of a virtual magnon, which then decays into a magnon and a phonon on account of interaction (4.3). The result is a real process with production of one magnon and one phonon, but now without equality of the momentum of the two quasiparticles. This increases greatly the final phase volume in comparison with the case (5.8). The corresponding expression for the decay probability, determined in second-order perturbation theory, is

$$W\approx I(I+1)\Phi_0\left(\frac{2A}{Z\Delta_0}\right)^2\left(\frac{\hbar\omega_D}{Z\Delta_0}\right)^{7/2}\frac{Z\Delta_0}{\hbar}. \qquad (5.12)$$

An estimate of this expression for alkali-atom crystals leads unexpectedly to a noticeable magnitude of the decay probability. The corresponding value of τ fluctuates in the interval $10-10^3$ sec and is different for different isotopes. This decay channel is not suppressed by an external magnetic field. In principle, however, it can be suppressed if in addition to electron polarization one can ensure polarization of the nuclear spins.

6. CONCLUSION

As shown by the foregoing analysis, good excitations in metastable polarized crystals are unique spin waves that carry negative energy. It is precisely the excitation of these spin waves, particularly simultaneously with phonons, which determines the principal channels of the decay of the metastable crystal. By using an external magnetic field of direction opposite to the polarization of the electron magnetic moments it is possible to suppress completely the single-magnon and two-magnon decay without phonon excitation or with excitation of one phonon, on account of the dipole–dipole interaction (see Sec. 5). The leading channels of the decay, which are not eliminated by the magnetic field, turn out to be connected with the decay of the polarized state into one magnon and two phonons via the dipole–dipole interaction, and into one magnon and one phonon via a second-order process connected simultaneously with the hyperfine interaction and with the magnon–phonon interaction of exchange origin. In the case of polarized crystals of alkali atoms, the first of them leads to a decay probability per particle $\sim 10^{-3}-10^{-4}$ sec^{-1}, and determines by the same token a rather large lifetime. The second channel turns out to be relatively faster, with a characteristic decay probability in the interval $\sim 10^{-1}-10^{-3}$ sec^{-1}. This probability varies substantially from element to element and from isotope to isotope, so that a suitable optimal selection can be made for experimental purposes. It must be emphasized that in the case of simultaneous polarization of the nuclear spins this decay channel can also be suppressed.

We have considered also another limiting case, corresponding to local spin flip with jarring of the phonon subsystem. As shown in Sec. 3, the probability of this decay is usually small compared with the value of W given above. We note that for polarized crystals of the type considered in the present paper, the situation corresponding to local spin flip is apparently not realized because of the large width of the spin-wave band compared with the Debye energy of the phonons.

[1)] In Ref. 1 the parameters of the polarized alkali-atom crystals were obtained on the basis of asymptotic interatomic-interaction potentials. A subsequent analysis has shown that this approach yields in reality only an order-of-magnitude estimate.

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