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Two-frequency microwave resonance in the hyperfine structure of the ground state of alkali metals

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The problem of two-particle resonance in a three-level system is considered. A stationary solution is obtained for the coherence of two levels coupled by alternating fields with the third; this solution is valid for arbitrary field intensities and frequencies. Two-frequency resonance was investigated experimentally in the hyperfine structure of the ground state of potassium under spectrally selected optical pumping. Modulation of the pump light registered transversely to the magnetic field through a circular analyzer was observed at a frequency equal to the difference between the two microwave fields that cause transitions between the level $F = 1$, $m_F = 1$ and the levels $F = 2$, $m_F = 1, 2$.

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1. INTRODUCTION

It is known that in magnetic-resonance techniques one uses two types of observable quantities, proportional to the imaginary and real components of the magnetic susceptibility. The former are connected with the populations of the magnetic sublevels and are responsible for energy absorption. The latter are connected with coherences, i.e., are described by the off-diagonal elements of the density matrix. Examples of these quantities are the oscillating (transverse) components of the magnetization. The populations and the coherences behave essentially differently at resonance. This is manifest by a different dependence of these quantities on the amplitude and the frequency of the alternating field and, frequently more importantly, in a difference in the dynamics. The registration of the quantities connected with the coherence is more convenient because the corresponding signals vary in time. Under stationary conditions they are expressed in the general case by a series in harmonics of the alternating fields that initiate the resonance. This makes it possible to register the resonance without resorting to a scanning technique, the use of which makes the dynamics of the observation worse and difficult in the case of very narrow resonance. In contrast to the populations, the time of establishment of which is limited by the relaxation constant, the phases of the coherence oscillations react without delay to changes in the system energy.

The advantages of observing the coherence can be realized in the region of relatively low frequencies—in the NMR technique[1] and in experiments on optical orientation of atoms in weak and medium fields.[2,3] Among the applications are the so called M_x-self-generating magnetometers with optical orientation, in which use is made of Larmor polarized-light precession due to the coherence of the atomic magnetic sublevels. The extension of this magnetic-resonance registration method to the region of higher frequencies encounters great technical difficulties. When optical means are used to monitor the coherence, the limitations are imposed primarily by the inertia of the photoreceivers. These limitations could be overcome in demonstration experiments,[4,5] in which microwave modulation of the light passing through potassium and rubidium vapor was observed under conditions of magnetic resonance in the hyperfine structure.

In this article we demonstrate the possibility of converting the microwave oscillations of coherence into radio-frequency oscillations by producing two-frequency microwave resonance on adjacent sublevels of the magnetic splitting of the hyperfine-structure states. The foregoing is illustrated in Fig. 1, which shows the energy structure of the ground state of an alkali-metal atom with nuclear spin 3/2 in a magnetic field. Assume that there exists a process that leads to predominant population of, say, the upper hyperfine level of $F = 1$. (This is a natural result of thermal relaxation, but the obtained population difference is very small and in practice one uses the forcing process of optimal pumping, see below). Superposition of two microwave fields of suitable polarization and frequency gives rise to transitions between one sublevel of the state $F = 1$ and two sublevels of $F = 2$, as is shown, for example, by the arrows of Fig. 1. Under these conditions, coherence sets in between the upper pair of levels at a frequency equal to the difference frequency of the two microwave fields.

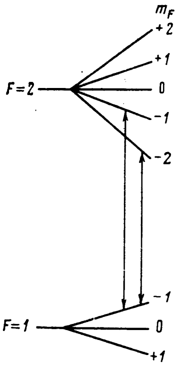

FIG. 1. Level scheme of the ground state of an alkali-metal atom with nuclear moment $I = 3/2$ in a magnetic field.

This coherence can be registered in the form of modulation of the light that causes transitions from an $F = 2$ level of the ground state to any one of the excited states (not shown in the figure). A two-frequency coherence resonance of this kind has a number of features that are explained in the next section.

2. CALCULATION OF TWO-FREQUENCY RESONANCE

The theoretical model of the situation shown in Fig. 1 is a three-level system with two variable fields. The problem of a three-level system in a strong field is the subject of an extensive literature, reference to which can be found in the monographs of Fain and Khanin[6] and of Letokhov and Chebotaev.[7] This problem arises in maser theory and in laser spectroscopy. Without confining ourselves to the usual approximation of the relative smallness of one of the fields, we obtain a general stationary solution for the case of interest to us. We introduce the notation for the levels, field frequencies (thick arrows) and transitions frequencies (thin arrows) indicated in Fig. 2. The purpose of the analysis is to determine the dependence of the density-matrix element $\rho_{23} = \rho^*_{32}$ on the detunings Δ_{ik} and on the field intensities. We use the equation[6,7]:

$$i\hbar\dot{\hat{\rho}} = [\hat{H}_0 + \hat{V}, \hat{\rho}] - [\hat{\Gamma}, \hat{\rho}], \tag{1}$$

where $\hat{H}_0$ is the Hamiltonian of the unperturbed system and $\hat{V}$ is the operator of the interaction with the external alternating fields. The phenomenological operator $\hat{\Gamma}$ describes the relaxation processes and is given by the matrix

$$(\hat{\Gamma}\hat{\rho})_{ik} = \begin{pmatrix} \gamma_{11}(\rho_{11}-\rho_{11}{}^0) & \gamma_{12}\rho_{12} & \gamma_{13}\rho_{13} \\ \gamma_{21}\rho_{21} & \gamma_{22}(\rho_{22}-\rho_{22}{}^0) & \gamma_{23}\rho_{23} \\ \gamma_{31}\rho_{31} & \gamma_{32}\rho_{32} & \gamma_{33}(\rho_{33}-\rho_{33}{}^0) \end{pmatrix}. \tag{2}$$

The real object of the investigation is the system of Fig. 1, subjected to optical pumping that ensures, in the absence of alternating fields, a stationary distribution of the populations ρ^0_{kk}. It is assumed that the pumping does not introduce any coherence, i.e., $\rho^0_{ik} = 0$. We seek the off-diagonal element in the form of complex harmonics that are in synchronism with the applied field:

$$\rho_{ik} = \tilde{\rho}_{ik} \exp\{-i\Omega_{ik}t\}. \tag{3}$$

It is assumed here that the matrix elements of the perturbation have an analogous time dependence:

$$V_{13}(t) = \tilde{V}_{13} \exp\{-i\Omega_{13}t\}, \quad V_{12}(t) = \tilde{V}_{12} \exp\{-i\Omega_{12}t\}. \tag{4}$$

This notation, which corresponds to the usual rotating-field approximation, presupposes that each of the two fields acts only on "its own" transition. This is easy to effect in practice by correctly choosing their polarizations. If this condition is not satisfied, then the solution remains valid near the resonances, i.e., at $\Delta_{ik} \ll \omega_{23}$.

Substitution of (2), (3), and (4) in (1) leads to a system of nine differential equations with constant coefficients, which goes over in the stationary case of steady-state populations ρ_{kk} and coherence amplitudes $\tilde{\rho}_{ik}$ into a system of nine algebraic equations. The general solution for the element $\tilde{\rho}_{23}$ of interest to us is quite cumbersome. Bearing in mind a concrete experiment, we present an expression for $\tilde{\rho}_{23}$ under the following simplifications. 1) The population differences $\rho^0_{33} - \rho^0_{11}$ and $\rho^0_{22} - \rho^0_{11}$ are equal, i.e., $\rho^0_{22} - \rho^0_{33} = 0$. This condition is satisfied exactly in the case of spectrally selective pumping with unpolarized light, which introduces a population difference only between sublevels pertaining to states with different total angular momentum F. 2) All the relaxation constants are equal to one another, $\gamma_{ik} = \gamma$. Under our conditions, this equality was satisfied within several dozen percent. We have

$$\tilde{\rho}_{23} = (\rho_{33}{}^0 - \rho_{11}{}^0)\, i\gamma V_2 V_3 \times \frac{A + B(V_2{}^2 + V_3{}^2) + 2i(V_2{}^2 + V_3{}^2)^2}{C + DV_2{}^2 + D'V_3{}^2 + EV_2{}^2V_3{}^2 + FV_2{}^4 + F'V_3{}^4 - 12\gamma(V_2{}^4V_3{}^2 + V_2{}^2V_3{}^4) + 4\gamma(V_2{}^6 + V_3{}^6)}. \tag{5}$$

We used the notation: $V_k = \tilde{V}_{1k}/\hbar = \tilde{V}_{k1}/\hbar$,

$$a_2 = \Delta_{21} - i\gamma = \omega_{21} - \Omega_{21} - i\gamma, \quad a_3 = \Delta_{13} - i\gamma = \omega_{13} - \Omega_{13} - i\gamma,$$
$$b = \Delta_{23} - i\gamma = \omega_{23} - \Omega_{23} - i\gamma, \; A = ia_2{}^*a_3{}^*b^*(a_2 + a_3),$$
$$B = i[a_3{}^*b^* - a_2{}^*(a_3 + a_2)], \; C = -\gamma|a_2a_3b|^2,$$
$$D = 2\gamma|a_2|^2 \operatorname{Re}(a_3b) + 4\gamma|a_3b|^2,$$
$$E = -2\gamma \operatorname{Re}(a_2{}^*a_3) + 4[\gamma(|a_2|^2 + |a_3|^2) + \operatorname{Im} a_2a_3b] + 12|b|^2\gamma, \quad F = -8\gamma \operatorname{Re}(a_3b) - \gamma|a_2|^2.$$

The quantities D' and F' are obtained from the unprimed quantities by interchanging the indices 2 and 3.

Let us point out the following features of the obtained solution.

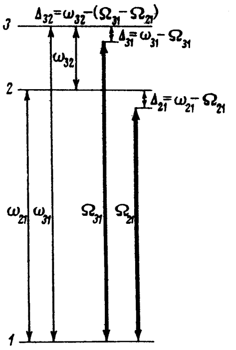

FIG. 2. Three-level system with the field frequencies (thick arrows) and transition frequencies (thin arrows) marked.

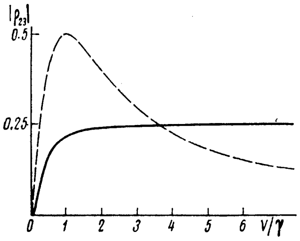

FIG. 3. Dependence of the coherence (in units of $\rho_{33}^0-\rho_{11}^0=\rho_{22}^0-\rho_{11}^0$) on the field intensity $V_2=V_3=V$ in units of γ. The dashed curve shows the variation of the coherence $\tilde{\rho}_{12}$ at resonance in a two-level system in units of $\rho_{22}^0-\rho_{11}^0$.

1) When $V_2=V_3=V$, the coherence of $\tilde{\rho}_{23}$ increases monotonically with increasing V, and approaches asymptotically a value that depends on the ratio of the relaxation constants. When all the constants are equal, this quantity is equal to $\frac{1}{4}(\rho_{33}^0-\rho_{11}^0)$. Under conditions of double resonance $\Delta_{12}=0$ and $\Delta_{13}=0$ the dependence of ρ_{23} on the perturbation $V_2=V_3=V$ takes the form

$$\tilde{\rho}_{23}=2V^2(\rho_{33}{}^0-\rho_{11}{}^0)(\gamma^2+8V^2)^{-1}, \tag{6}$$

which is plotted in Fig. 3. This variation of the coherence differs substantially from the behavior of the coherence in a two-level system, where it goes through a maximum with increasing intensity of the alternating field and then tends to zero, as shown for comparison dashed in Fig. 3.

2) The phase φ of the coherence $\tilde{\rho}_{23}$ in the resonance $\Delta_{23}=0$ does not depend on either the field intensities V_2 and V_3 or on the detuning $\Delta_{13}=-\Delta_{23}$. In other words, if the difference of the frequencies of the two fields V_2 and V_3 coincides with the transition frequency Ω_{23}, then the phase φ is constant. This makes it possible to fix the resonance of the frequency difference $\Delta_{23}=0$ without having to worry about the stability of the frequencies Ω_{12} and Ω_{13} themselves. In contrast to single-particle resonance in a two-level system, however, the slope of the phase characteristic $\partial\varphi/\partial\Delta_{23}$ depends on the intensity of the alternating field.

Figure 4 shows the dependence of the slope $\partial\varphi/\partial\Delta_{23}$ at the resonance $\Delta_{23}=0$ for $\Delta_{13}=0$ and $V_2=V_3=V$ in accordance with the expression

$$\partial\varphi/\partial\Delta_{23}=-{}^3/{}_2\gamma(\gamma^2+2V^2)^{-1}. \tag{7}$$

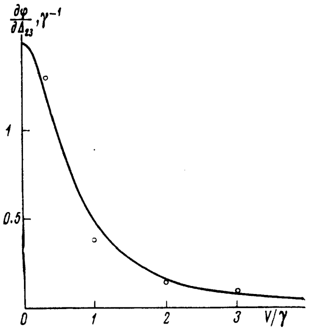

FIG. 4. Slope of the phase characteristic of $\tilde{\rho}_{23}$ as a function of the field intensities $V_2=V_3=V$ in units of γ. The experimental points are marked on the curve.

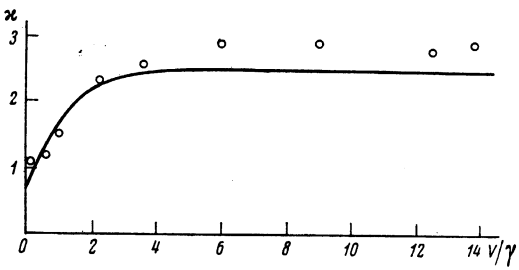

FIG. 5. Ratio $\varkappa$ of the widths of the resonances with $\Delta_{23}=0$ and with $\Delta_{13}=0$ as a function of the fields $V_2=V_3=V$ in units of γ. Solid line—calculation.

3) By virtue of the two-frequency character of the resonance, it is possible to go through it in two ways: by fixing one of the resonance, for example, $\Delta_{13}=0$ and varying the frequency of the second field, or by fixing the resonance of the frequency difference $\Delta_{23}=0$ and varying in like fashion the frequencies Ω_{13} and Ω_{12}. By registering in this case, for example, the modulus of $\tilde{\rho}_{23}$, we obtain two different families of resonances that are differently broadened by the alternating fields. The resonances obtained under the condition $\Delta_{23}=0$ are more strongly broadened. This is shown in Fig. 5, where the solid line represents the ratio $\varkappa$ of the widths at half the frequency of the resonances of these two types as a function of the intensity of the alternating field for the case $V_2=V_3=V$. At high intensities, the widths of the resonances increase linearly with increasing V and retain a constant ratio.

In concluding this section, we point out the connection between ρ_{23} and the experimentally observed absorption of polarized light. The spectral density k_ν of the absorption coefficient of an atomic vapor is given by

$$k_\nu=c\sum_{\mu,\mu',m}\rho_{\mu\mu'}\langle m|\hat{\mathbf{d}}\mathbf{e}|\mu\rangle\langle\mu'|\hat{\mathbf{d}}\mathbf{e}|m\rangle C_m(\nu), \tag{8}$$

where c is a proportionality coefficient, $\langle m|\hat{\mathbf{d}}\mathbf{e}|\mu\rangle$ is the matrix element of the dipolar transition from the ground state with magnetic sublevels into an excited state with sublevels m, $\mathbf{e}$ is the polarization vector, and $C_m(\nu)$ is the contour of the absorption line on the transition $\mu-m$.

3. EXPERIMENT

K^{39} vapor saturated at 60 °C, with argon (10 Torr) as the buffer gas, was subjected to spectrally selected pumping by resonant radiation at 769.9 and 766.5 nm, filtered by K^{41} vapor. As a result of the filtering, the center of gravity of each of the lines is shifted relative to the absorption lines of K^{39}, leading to a predominant excitation of atoms from the $F=2$ level of ground state.[8,9] The results of the selective excitation is enrichment of the state $F=1$, which is accompanied by a decrease of the optical absorption. Population changes due to the induced transitions between the hyperfine states are detected optically by the increased absorption.

Out of the entire spectrum of the microwave transitions of potassium, which contained nine lines, the three-level model considered above agrees with the following combinations: the level 1 is taken to be $F=1$, $m_F=\pm 1$, and the role of the levels 2 and 3 can be assumed by the levles $F=2$, $m_F=\pm 1, \pm 2$ or of the levels $F=2$, $m_F=0, \pm 2$. The conditions for the registration of coherence in these two cases are different. They can be established from an analysis of expression (8), which should determine the optimal polarization of the controlling light and its spectral composition, since these determine the contribution made to the signal by different excited levels m. The latter are the sublevels of the fine and hyperfine structure of the first excited state $4\,^2P_{1/2,3/2}$. The question of the conditions of registration is easiest to solve by changing over from $\rho_{\mu\mu'}$ to the components of the statistical tensor $\rho_q^\varkappa$ in accordance with the scheme summarized by Chaĭka.[10] Without dwelling on the calculations, we present a summary of the results.

The coherence of the sublevels $m_F=0$ and $m_F=\pm 2$ is best registered in absorption of light that is linearly polarized perpendicular to the magnetic-field vector, under the mandatory condition that the quantities $C_m(\nu)$ in (8) pertaining to different levels of the hyperfine splitting of the excited state be different. This requirement is not satisfied in the case of potassium, for which the difference is much less than the Doppler line width. Therefore, no interference should be observed between the levels $m_F=0$ and $m_F=\pm 2$.

The coherence of the levels $m_F=1$ and $m_F=2$ is best observed in absorption of light having circular polarization and directed transverse to the magnetic field. The necessary condition here is a difference between the intensities of the hyperfine-splitting components 769.9 and 766.5 nm, which is easily obtained. In connection with the foregoing, we chose the following experimental setup. The pump light was directed across the magnetic field, and after going through a cell with potassium vapor it passed through a filter that separated the 769.9 nm line and through a circular analyzer mounted in front of the photoreceiver. The experiments were performed in a magnetic field 0.02 Oe produced by a solenoid inside a magnetic screen. The corresponding of the levels 2 and 3 was 14 kHz. The fields V_2 and V_3 were produced by an inductor fed from two independent frequency synthesizers through a power amplifier. The field frequencies Ω_{12} and Ω_{13} could be tuned in discrete fashion in steps of 1 Hz in the vicinity of the hyperfine-splitting of the ground state of potassium, equal to 461.7 MHz. Each of the frequencies Ω_{12} and Ω_{13} could be tuned independently to its own resonance. The quantities V_2 and V_3 were calibrated against the broadening of the lines of single microwave resonances. A preliminary investigation has shown that the widths of the single-frequency microwave resonances and of the radiofrequency resonance between the sublevels of the magnetic splitting of the hyperfine component (which was observed by using an optical orientation technique) are determined principally by the optical broadening. The widths of these resonances at half height turned out to be equal within 20% at approximately 150 Hz.

Under the described conditions, we observed a powerful pump-light modulation signal at the frequency Ω_{23}. Its value was compared with the signals due to the change of the absorption of the potassium under the conditions of single-frequency microwave resonance and was equal to as much as 10% of the latter, as against the calculated 25%. This discrepancy is attributed to the insufficiently complete separation of the components of the resonance doublet in the registered radiation. We checked on the following predictions of the theory:

1) Independence of the depth of modulation of the light of the intensity V in the saturation region. In accordance with the plot of Fig. 3, a rapid growth of the signal at the frequency Ω_{23} is observed with increasing V, followed by saturation. In the experiment, the parameter V/γ ranged from zero to 15. Loss of equality of the perturbations V_2 and V_3 in saturation region of the signal caused it to decrease in accordance with (5).

2) A comparison of the width of the resonances under the condition $\Delta_{23}=0$ and $\Delta_{13}=0$ revealed good agreement with the calculation, as illustrated in Fig. 5.

3) We investigated the phase chracteristics of the resonances. We used for this purpose a phase meter, the reference voltage for which was produced by subtracting the frequencies Ω_{13} and Ω_{12}. It was confirmed that if $\Delta_{23}=0$ the phase of the modulation at the frequency Ω_{23} is independent of the detuning $\Delta_{13}=-\Delta_{21}$ of the frequencies Ω_{13} and Ω_{21} in the entire region where the signal amplitude was high enough to be registered. At a fixed frequency of one of the fields ($\Delta_{13}=0$), variation of the frequency of the other led to a linear change of the light-modulation phase in the vicinity of the resonance. The slope of the phase characteristic was dependent in this case on the intensity of the alternating field, in agreement with the calculation (see Fig. 4).

4) An attempt was made to observe coherence of the resonances of the states $F=2$, $m_F=0$ and 2. To this end, the circular analyzer in front of the photodetector was replaced by a linear one and the microwave-field frequencies were chosen in suitable fashion. No modulation signal was observed, accurate to 1% of the maximum signal of the coherence of the sublevels $m_F=1$ and 2.

We note in conclusion that we observed also some deviations from the theory. Although the ratio of the widths of the resonances $\Delta_{23}=0$ and $\Delta_{13}=0$ agree with the calculation, the absolute values of these widths turned out to be approximately half those expected in the region $V \gg \gamma$. This discrepancy may be the result of the simplified description of the relaxation processes.

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Theory of the absorption band shape and the fluorescence band shape of an impurity center in the Condon approximation

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We study the effect of the electron-phonon interaction $\hat{W}$, quadratic in the phonon operators, on the shape of the optical absorption and fluorescence bands of an impurity center. We do not assume that $\hat{W}$ is small. By solving an integral equation we obtain analytical expressions for the phonon functions which are responsible for the occurrence of phonon wings (PW) in the optical spectrum. We evaluate numerically on a computer the above-mentioned phonon functions. We analyze the breaking of the mirror symmetry of the absorption and fluorescence spectra, caused by the interaction $\hat{W}$, and also the effect of $\hat{W}$ on the integral intensity of the non-phonon line (NPL). We establish a simple analytical connection between the temperature broadening of the NPL and the above-mentioned lack of mirror symmetry of the phonon functions.

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INTRODUCTION

The intensity distribution, and also a number of important details of the optical band that corresponds to an allowed electron transition, can very often be explained in the Condon approximation. In that approximation the electron-phonon interaction is the difference $\hat{\Lambda}$ of the adiabatic potentials of the excited and the unexcited crystals. Expanding $\hat{\Lambda}$ in terms of the deviations of the molecules from their equilibrium positions we get the interaction $\hat{V}$ which is linear in the deviations and the interaction $\hat{W}$ which is quadratic in them.

So far the role of the linear interaction has been studied in great detail.[1–8] Appreciably less attention has been paid to the effect of the quadratic interaction $\hat{W}$ on the optical band shape. It is known that $\hat{W}$ may be responsible for the temperature broadening and the shift of the non-phonon lines (NPL)[9,10] and that it can also break down the mirror symmetry of the absorption and fluorescence bands.[11] The quadratic interaction therefore produces effects which are very often encountered experimentally. However, these effects may also be produced by other causes. It is therefore sometimes impossible to indicate exactly the cause of an observed effect without a quantitative comparison of theory and experiment. Unfortunately, it is difficult to compare quantitatively the formulae of[9–11] with experiments as perturbation theory was used in[9,10] to consider the quadratic interaction and the criteria for the applicability of the formulae given was not indicated, while in[11] such characteristics of the bands as the second moment which give little information were studied. It is true that Kubo and Toyazawa[6] took the effect of the linear and the quadratic interactions on the absorption band shape rigorously into account, but their formulae are very complicated and do not give a practical solution of the problem.

Recently, Levenson[12] and, independently, the present author[13] have proposed a different way to take the effect of $\hat{W}$ on the band shape into account. In their approach it is necessary for the evaluation of the band shape to find a function which is a solution of an integral equation. By solving this equation in various particular cases it was possible to get a number of new results referring to NPL and the phonon wing (PW).[13–15] However, a number of problems remained unanswered as we were not able to find an effective solution of the above-mentioned integral equation in the general case. We find such a solution in the present paper. Using it we obtain in Secs. 4 and 5 a few new physical results.

1. INTERACTION FUNCTION

The vibrational Hamiltonian $H^g(\mathbf{R})$ of a crystal with an unexcited ($g=0$) and with an excited ($g=e$) impurity can be written as follows:

$$H^g(\mathbf{R}+\mathbf{a}^g)={}^1/_2(\dot{\mathbf{R}}^2+(\mathbf{R}+\mathbf{a}^g)U^g(\mathbf{R}+\mathbf{a}^g)).$$

The coordinates R_q of the molecule are connected with the normal dimensionless coordinates $R^g_\varkappa$ of the crystal through the transformation1)

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