TABLE V. Comparison of the γ values of the first superlattice reflections in two states with order parameter $S>0.9$ and $S=0.6$, respectively.

(hkl)	$S>0.9$ $-\gamma$	$S=0.6$ $-\gamma$
(100)	0.460 ± 0.009	0.400 ± 0.018
(110)	0.419 ± 0.005	0.331 ± 0.019
(210)	0.317 ± 0.006	0.193 ± 0.022
(211)	0.299 ± 0.004	$0.24\ \pm0.04$
(221)	0.229 ± 0.007	0.141 ± 0.017

superlattice peaks with the state of order (Table V) rules out such an hypothesis, which would imply that the magnetic moments on Co and Pt in the ordered matrix should stay constant even if the average order parameter decreased.

Finally, the magnetic-density distribution indicates a fair agreement with calculations as far as the spherical part is concerned. The fact that the cobalt form factor in $CoPt_3$ lies a little lower than the one measured by Moon in the pure metal[19] could possibly be ascribed to a different exchange-polarization effect and orbital contribution.

In regard to the aspherical part, we have seen that the symmetry of the unpaired $3d$ electrons of cobalt in this alloy is mainly t_{2g} independently of the order, in contrast with the predominantly e_g symmetry of magnetic electrons in fcc Co.[27] It has been noted by Shirane *et al.*[3] that in the Pd-Fe system there is a trend from e_g to t_{2g} symmetry as the Fe content is decreased; the results on Co and $CoPt_3$ seem to show a similar trend in regard to the Co content. In fact some preliminary results on a CoPt disordered alloy[29] indicate that the symmetry, though of t_{2g} type, is less prominent than in $CoPt_3$.

ACKNOWLEDGMENTS

The first part[10] of this work was done with the essential cooperation of Dr. G. Mazzone to whom we are very grateful. We acknowledge also the participation of Dr. M. Mirabile, who attended to part of the measurements on the disordered and partially ordered samples while preparing his thesis.

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[29] B. Antonini, F. Menzinger, and A. Paoletti (unpublished).

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Interaction between Light Waves and Spin Waves*

Y. R. SHEN
Department of Physics, University of California, Berkeley, California

AND

N. BLOEMBERGEN†
Department of Electrical Engineering, University of California, Berkeley, California

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The coupling of photons and magnons can be treated by the same methods developed for the coupling between photons and phonons. The coupled wave equations are derived directly from the Hamiltonian density for the quantized fields with the density-matrix formalism. The similarity between the spin Raman effect and the vibrational Raman effect is emphasized and it is shown that the spin Raman effect will usually be one or two orders of magnitude smaller than the vibrational effect in Raman liquids. The possibility of exiciting spin-wave modes by light in ferro-, ferri-, and antiferromagnetic materials is discussed. The combined coupling of magnetic, vibrational, and light waves is also analyzed and a magnon excitation may be induced by the stimulated Brillouin effect on a magnetoelastic mode.

I. INTRODUCTION

THE Raman effect can be described as a second-order inelastic scattering of light, in which the scattering system makes a transition to an excited state.[1] Originally the spontaneous Raman scattering was almost exclusively employed to study vibrational and rotational excitations of molecules.[2] Loudon[3] suggested that electronic excitations of transition-metal ions should be observable in the Raman effect. Hougen and Singh[4] independently succeeded in finding this purely electronic Raman effect for Pr^{3+} ions in LaF_3.

It is also possible for the excitation to be of a purely

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† On leave from Harvard University.

[1] P. A. M. Dirac, Proc. Roy. Soc. (London) A114, 710 (1927).

[2] See, for example, G. Placzek, *Marx Handbuch der Radiologie*, edited by E. Marx (Academische Verlagsgesellschaft, Leipzig, Germany, 1934), 2nd ed., Vol. VI, part II, p. 209.

[3] R. J. Elliott and R. Loudon, Phys. Letters 3, 189 (1964); R. Loudon, Advan. Phys. 13, 423 (1964).

[4] J. T. Hougen and S. Singh, Phys. Rev. Letters 10, 406 (1963).

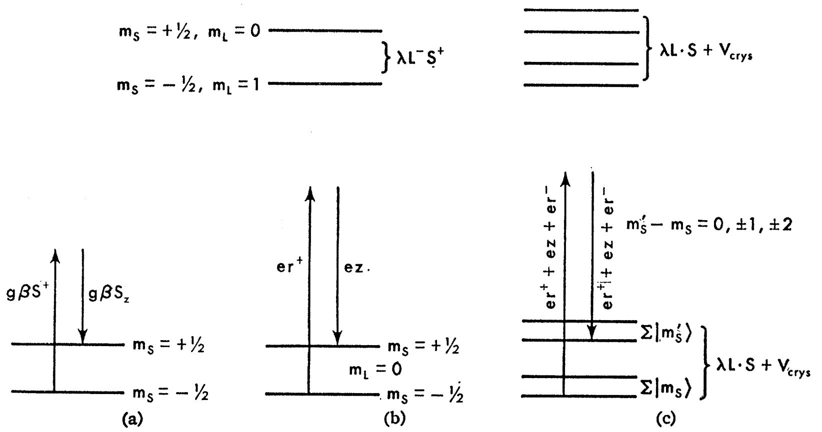

FIG. 1. Energy diagrams showing the spin-Raman processes. (a) Magnetic-dipole Raman transitions in a two-level spin system; (b) electric-dipole Raman transitions in a system with negligible crystalline field; (c) electric-dipole Raman transitions in a system with an appreciable crystalline field.

magnetic nature. In this case the excited state differs from the ground state in the spin magnetic quantum number. Such two-photon processes are well known in magnetic resonance.[5] The Raman susceptibility for the purely magnetic dipole transitions in a two-level system [Fig. 1(a)] has been reviewed by the present authors.[6] The final state with a different magnetic quantum number may also be reached with electric dipole transitions via a virtual electronic excited state, as shown in Fig. 1(b). It is of course necessary in this case to invoke spin-orbit coupling to change the spin quantum number. This process was also suggested by Loudon.[3] In the simplest case, the transition from the state $m_s=-\frac{1}{2}$ to $m_s=+\frac{1}{2}$ in a Kramers ground-state doublet of a $^2S_{1/2}$ ion would take place with a virtual optical transition to the $^2P_{3/2}$ or $^2P_{1/2}$ manifold. Although the optical electric dipole matrix elements cannot change the spin magnetic quantum number, it is possible to reach the final state with $\Delta m_s=\pm1$ by invoking the spin-orbit coupling, as indicated in Fig. 1(b). More generally, for transition metal ions with spin-orbit coupling in crystalline fields of arbitrary strength, different magnetic sublevels of the ground-state multiplet could be reached via a two-photon process with electric dipole matrix elements, provided the initial and final states have components whose magnetic quantum numbers differ by $\Delta m_s=0$, ±1 or ±2, as shown in Fig. 1(c).

In ferro-, ferri-, and antiferromagnetic materials, the spin excitation is not localized and the elementary excitation is described as a spin wave. It is the purpose of this paper to present the formalism which describes the coupling of light waves to these spin-wave excitations and to discuss the possibility of observing the stimulated spin Raman effect in magnetic media.

The same formalism that was developed[7,8] to describe the coupling of light waves with acoustical waves (stimulated Brillouin effect) and with optical phonons (stimulated Raman effect) can be adapted to the case of magnetic excitations. In Sec. II, the general quantum mechanical formulation for coupled boson fields is applied to the coupling of electromagnetic fields and vibrations. The wave equations for the expectation values of the fields and the nonlinear coupling constants are derived directly from a Hamiltonian. Although localized electronic states are used, as would be appropriate for insulators, the considerations could readily be extended to conductors by using itinerant Bloch wave functions.

In Sec. III this same procedure is applied to magnons. The exponential gain for the Stokes wave in the spin Raman effect is derived from the coupled wave equations. The result reduces to that derived from a spin Raman susceptibility for isolated magnetic ions, which would be appropriate in the paramagnetic case.

The possibility of detecting the spin Raman effect in various magnetic systems is discussed in Sec. IV. Some explicit equations are given for the two-sublattice model for ferri- and antiferromagnetic materials. The spin Raman effect is roughly 1 or 2 orders of magnitude smaller than the ordinary Raman effect in liquids, because the oscillator strengths of the electronic transitions involved in the magnetic ions are smaller than those involved in the molecules. In Sec. V, the general case of coupling between laser, Stokes, and infrared electromagnetic waves with phonon and magnon waves is discussed. A magnetic excitation could be induced by the combination of the stimulated Brillouin and the spin Raman effect.

II. COUPLING OF LIGHT WITH PHONONS

A detailed calculation of the ordinary stimulated Raman and Brillouin scattering has been given earlier.[8] In this section, a brief review of the subject is given in order to develop notations convenient for the later discussion of coupling of light with magnons. This also affords the opportunity to generalize the formalism so

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[5] A. Javan, J. Phys. Radium **19**, 836 (1958); J. M. Winter, *ibid.* **19**, 834 (1958).

[6] N. Bloembergen and Y. R. Shen, Phys. Rev. **133**, A37 (1964).

[7] E. Garmine, F. Pandarese, and C. H. Townes, Phys. Rev. Letters **11**, 160 (1963); R. W. Hellwarth, Current Sci. India **33**, 129 (1964).

[8] N. Bloembergen and Y. R. Shen, Phys. Rev. Letters **12**, 504 (1964). Y. R. Shen and N. Bloembergen, Phys. Rev. **137**, A1787 (1965).

that both the light and the other coupled boson fields are quantized. The wave equations together with the coupling constants are derived directly from the total Hamiltonian of the system

$$\mathcal{H}=\mathcal{H}_{\mathrm{rad}}+\mathcal{H}_{\mathrm{phonon}}+\mathcal{H}_{\mathrm{elec}}+\mathcal{H}_{\mathrm{int}}. \tag{1}$$

The Hamiltonian for the radiation field can be quantized in the usual way,[9] as well as the phonon Hamiltonian in its harmonic approximation.[10]

$$\begin{aligned}\mathcal{H}_{\mathrm{rad}}&=\sum_{\mathbf{k},i}\hbar\omega_{\mathbf{k},i}(\alpha_{\mathbf{k}i}{}^{\dagger}\alpha_{\mathbf{k}i}+\tfrac{1}{2}),\\ \mathcal{H}_{\mathrm{phonon}}&=\sum_{\mathbf{q},j}\hbar\omega_{\mathbf{q},j}(a_{\mathbf{q}j}{}^{\dagger}a_{\mathbf{q}j}+\tfrac{1}{2}).\end{aligned} \tag{2}$$

α, $\alpha^\dagger$ and a, $a^\dagger$ are the annihilation and creation operators for photons and phonons, respectively. Their operations on the number states yield $\alpha|n\rangle=n^{1/2}|n-1\rangle$ and $\alpha^\dagger|n\rangle=(n+1)^{1/2}|n+1\rangle$. The photon and the phonon wave vectors are indicated by $\mathbf{k}$ and $\mathbf{q}$, respectively. The particular phonon branch under consideration is labeled by j.

The interaction Hamiltonian consists of two parts, the electron-phonon interaction and the electron-radiation interaction, which may be written in the form

$$\mathcal{H}_{\mathrm{int}}=\mathcal{H}_{\mathrm{e\text{-}p}}+\mathcal{H}_{\mathrm{e\text{-}r}}, \tag{3}$$

with

$$\begin{aligned}\mathcal{H}_{\mathrm{e\text{-}r}}&=-\sum_{m,b}(e\mathbf{r})_{mb}\cdot\mathbf{E}_{mb},\\ \mathcal{H}_{\mathrm{e\text{-}p}}&=\sum_{m,b}\sqrt{M_{mb}}\mathbf{U}_{mb}\cdot\mathbf{f}_{mb}.\end{aligned} \tag{4}$$

In a nonpolar medium the phonon-radiation interaction can be neglected. The Raman effect in polar media has been discussed elsewhere.[11] In Eq. (4), $e\mathbf{r}$, $\mathbf{E}$, M, $\mathbf{U}$, and $\mathbf{f}$ are the electric dipole, the electric field, the atomic mass, the atomic displacement, and the generalized force on the atom, respectively. The indices m and b refer to the bth atom in the mth unit cell of the lattice. Both operators $\mathbf{E}$ and $\mathbf{U}$ can be expanded in terms of annihilation and creation operators. In the Schrödinger representation,

$$\begin{aligned}\mathbf{E}_{mb}&=\textstyle\sum_{\mathbf{k}}[\mathbf{E}^+(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{R}_m}+\mathbf{E}^-(\mathbf{k})e^{-i\mathbf{k}\cdot\mathbf{R}_m}],\\ \mathbf{E}^+(\mathbf{k})&=i(2\pi\hbar\omega_{\mathbf{k}}/V)^{1/2}\hat{\epsilon}\alpha_{\mathbf{k}},\\ \mathbf{E}^-(\mathbf{k})&=i(2\pi\hbar\omega_{\mathbf{k}}/V)^{1/2}\hat{\epsilon}^*\alpha_{\mathbf{k}}{}^{\dagger},\end{aligned} \tag{5a}$$

where the fields are normalized with respect to a volume V;

$$\begin{aligned}\mathbf{U}_{mb}&=\sum_{\mathbf{q}\cdot j}[\mathbf{U}^+(\mathbf{q},b,j)e^{i\mathbf{q}\cdot\mathbf{R}_m}+\mathbf{U}^-(\mathbf{q},b,j)e^{-i\mathbf{q}\cdot\mathbf{R}_m}],\\ \mathbf{U}^+(\mathbf{q},b,j)&=(\hbar/2M_bN\omega_{qj})^{1/2}\mathbf{e}(\mathbf{q},b,j)a_{\mathbf{q}j},\\ \mathbf{U}^-(\mathbf{q},b,j)&=(\hbar/2M_bN\omega_{qj})^{1/2}\mathbf{e}^*(\mathbf{q},b,j)a_{\mathbf{q}j}{}^{\dagger},\\ \{\mathbf{e}(\mathbf{q},b,j)\}\cdot\{\mathbf{e}(\mathbf{q},b,j)\}^{\dagger}&\equiv\textstyle\sum_b\mathbf{e}(\mathbf{q},b,j)\cdot\mathbf{e}^*(\mathbf{q},b,j)=1.\end{aligned} \tag{5b}$$

Here N is the number of unit cells in the lattice, $\mathbf{R}_m$ the position of the mth unit cell, $\hat{\epsilon}$ the unit vector indicating polarization of the E field, and $\{e\}$ a set of b vectors with $\mathbf{e}(\mathbf{q},b,j)$ denoting the relative displacement of the bth atom in a unit cell corresponding to the phonon mode specified by $\mathbf{q}$ and j.

The wave equations for photons and phonons can be derived using the density-matrix formalism. Let ρ be the density-matrix operator for the entire system. The density-matrix operator for the radiation system alone is obtained by taking the trace over electron and phonon systems, such that $\rho_r=\mathrm{Tr}_{(ep)}\rho$. From the equation of motion for ρ, we find

$$\begin{aligned}&\left(\frac{\partial^2}{\partial t^2}+\omega_{\mathbf{k}}{}^2\right)\langle(n+1)_{\mathbf{k}}|\rho_r(t)|n_{\mathbf{k}}\rangle\\ &=\left(\frac{\partial}{\partial t}-i\omega_{\mathbf{k}}\right)\\ &\qquad\times(1/i\hbar)\,\mathrm{Tr}_{(\mathrm{e\text{-}p})}\langle(n+1)_{\mathbf{k}}|[\mathcal{H}_{\mathrm{int}},\rho(t)]|n_{\mathbf{k}}\rangle,\end{aligned} \tag{6}$$

where $\langle n_{\mathbf{k}}|$ is the photon number state for n photons in the mode $\mathbf{k}$. With $\langle\mathbf{E}_{\mathbf{k}}(\mathbf{R},t)\rangle=\mathrm{Tr}\rho(t)\mathbf{E}^+(\mathbf{k})\exp(i\mathbf{k}\cdot\mathbf{R})$ and $\omega_{\mathbf{k}}=kc$, and with the aid of Eqs. (2)–(5), the above equation yields the wave equation for $\langle\mathbf{E}_{\mathbf{k}}(\mathbf{R},t)\rangle$. In first approximation with $\rho(t)=\rho_r(t)\rho_{ep}(t)$, one finds

$$\left(\frac{1}{c^2}\frac{\partial^2}{\partial t^2}+k^2\right)\langle\mathbf{E}_{\mathbf{k}}(\mathbf{R},t)\rangle=-\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\langle\mathbf{P}_{\mathbf{k}}(\mathbf{R},t)\rangle, \tag{7}$$

where $\langle\mathbf{P}_{\mathbf{k}}(\mathbf{R},t)\rangle=\mathrm{Tr}\rho(t)e\mathbf{r}$ is proportional to $\exp(i\mathbf{k}\cdot\mathbf{R}-i\omega_{\mathbf{k}}t)$. In summing over all Fourier components, one can replace the factor k^2 by $-\nabla^2$; Eq. (7) then reduces to the classical wave equation for $\langle\mathbf{E}(\mathbf{R},t)\rangle$.

The radiation density as measured by photosensitive detectors is, however, proportional to $\langle|E^2|\rangle=\mathrm{Tr}\rho\mathbf{E}^+\mathbf{E}^-$. The differential equation for $\langle|E^2|\rangle$ can also be readily derived from the equation of motion for ρ. As expected, the spontaneous emission noise, if present will turn out in this full quantum-mechanical treatment. The noise problem in parametric quantum oscillators and amplifiers has been discussed by other authors in the Heisenberg representation.[12] In the classical treatment, the spontaneous emission noise can usually be taken into account in an *ad hoc* manner by inserting in the field amplitude equation a noise term with a random phase. In the following discussion, we are mainly interested in the parametric amplification of a coherent input field. The spontaneous noise will not be considered. The radiation fields will be treated classically, since the quantized field treatment yields exactly the same results as long as the approximation $\rho(t)=\rho_r(t)\rho_{ep}(t)$ is made.

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[9] See, for example, W. Heitler, *Quantum Theory of Radiation* (Oxford University Press, New York, 1954).

[10] See, for example, J. M. Ziman, *Electrons and Phonons* (Clarendon Press, Oxford, 1960).

[11] Y. R. Shen, Phys. Rev. **137**, A1741 (1965).

[12] W. H. Louisell, A. Yariv, and A. E. Siegman, Phys. Rev. **124**, 1646 (1961).

We shall assume the presence of only two em modes, such that

$$\mathbf{E}(\mathbf{R},t)=\boldsymbol{\mathcal{E}}_l\exp[i\mathbf{k}_l\cdot\mathbf{R}-i\omega_l t]+\boldsymbol{\mathcal{E}}_S\exp[i\mathbf{k}_S\cdot\mathbf{R}-i\omega_S t]+\text{complex conjugate}.$$

The phonon wave equation can be obtained from the equation of motion for ρ_{ep}. For simplicity, we assume only two electronic states for each atom, the ground state $\langle g|_{mb}$ and the excited state $\langle i|_{mb}$. Let $\langle G|=\prod_{m,b}\langle g_{mb}|$. The equation of motion for ρ_{ep} yields

$$\left(\frac{\partial^2}{\partial t^2}+2\Gamma\frac{\partial}{\partial t}+\omega_{\mathbf{q}}^2\right)\langle(n+1)_{\mathbf{q}},G|\rho_{ep}{}^{(\omega)}|n_{\mathbf{q}},G\rangle e^{-i\omega t}=(-1/h)\left(\omega_{\mathbf{q}}+i\frac{\partial}{\partial t}\right)\langle(n+1)_{\mathbf{q}},G|[\mathcal{H}_{\text{e-r}},\rho_{ep}]^{(\omega)}|n_{\mathbf{q}},G\rangle e^{-i\omega t},\tag{8}$$

where $\omega=\omega_l-\omega_S$, $n_{\mathbf{q}}$ denotes the number of phonons with wave vector $\mathbf{q}$ and frequency $\omega_{\mathbf{q}}$, and Γ is the phenomenological damping constant. The matrix element of $[\mathcal{H}_{\text{e-r}},\rho_{ep}]$ can be calculated by a perturbation expansion. The lowest order nonvanishing result is

$$\langle(n+1)_{\mathbf{q}},G|[\mathcal{H}_{\text{e-r}},\rho_{ep}]^{(\omega)}|n_{\mathbf{q}},G\rangle=\sum_{m,I}\left[\frac{\langle(n+1)_{\mathbf{q}},g|\sum_b(e\mathbf{r})_b\cdot\boldsymbol{\mathcal{E}}_S{}^*|I\rangle\langle I|\sum_b(e\mathbf{r})_b\cdot\boldsymbol{\mathcal{E}}_l|n_{\mathbf{q}},g\rangle}{\hbar(\omega_l-\omega_{I,gn_{\mathbf{q}}})}-\frac{\langle(n+1)_{\mathbf{q}},g|\sum_b(e\mathbf{r})_b\cdot\boldsymbol{\mathcal{E}}_l|I\rangle\langle I|\sum_b(e\mathbf{r})_b\cdot\boldsymbol{\mathcal{E}}_S{}^*|n_{\mathbf{q}},g\rangle}{\hbar(\omega_S+\omega_{I,gn_{\mathbf{q}}})}\right](\rho_{n_{\mathbf{q}}}{}^0-\rho_{(n+1)_{\mathbf{q}}}{}^0)\exp[i(\mathbf{k}_l-\mathbf{k}_S)\cdot\mathbf{R}_m].\tag{9}$$

Here $(\rho_{n_{\mathbf{q}}}{}^0-\rho_{(n+1)_{\mathbf{q}}}{}^0)$ is the average population difference between the phonon number states $\langle n_{\mathbf{q}}|$ and $\langle(n+1)_{\mathbf{q}}|$ at an arbitrary temperature and $\langle I|$ is the intermediate state with arbitrary mixing of electronic and vibrational character. The states $\langle(n+1)_{\mathbf{q}}|$ and $|n_{\mathbf{q}}\rangle$ in the square bracket yield a factor $\exp(-i\mathbf{q}\cdot\mathbf{R}_m)$ in the explicit calculation. The above matrix element is therefore nonvanishing only if the momentum matching condition $\mathbf{q}=\mathbf{k}_l-\mathbf{k}_S$ is satisfied, since otherwise $\sum_m\exp[i(\mathbf{k}_l-\mathbf{k}_S-\mathbf{q})\cdot\mathbf{R}_m]=0$. The matrix element would vanish if the electron-phonon interaction were absent, as the electron-radiation interaction cannot change the occupation number of phonons.

In the long-wavelength limit, the dispersion of the phonon modes has the form

$$\omega_{\mathbf{q}}^2=\omega_0^2+\beta q^2.$$

For acoustic phonons, β is positive and $\omega_0=0$. The phonon wave of wave vector $\mathbf{q}$ attached to the ground electronic state $\langle g|$ can be defined in terms of a dimensionless normal coordinate,

$$\begin{aligned}\langle\mathbf{Q}_{\mathbf{q}}(\mathbf{g},\mathbf{R},t)\rangle&=\langle G|\textstyle\sum_b(2M_b\omega_{\mathbf{q}}/\hbar)^{1/2}\operatorname{Tr}_{(p)}\rho_{ep}{}^{(\omega)}\mathbf{U}^+(q,b)\exp(i\mathbf{q}\cdot\mathbf{R}-i\omega t)|G\rangle\\&=\langle\mathbf{Q}(\mathbf{q},\omega)\rangle\exp(i\mathbf{q}\cdot\mathbf{R}-i\omega t).\end{aligned}\tag{10}$$

Equations (9) and (10) lead to the phonon wave equation

$$\left[\frac{\partial^2}{\partial t^2}+2\Gamma\frac{\partial}{\partial t}+\omega_0^2-\beta\nabla^2\right]\langle Q_{\mathbf{q}}(g,\mathbf{R},t)\rangle=\boldsymbol{\lambda}:\mathbf{E}_l\mathbf{E}_S{}^*\exp(i\mathbf{q}\cdot\mathbf{R}-i\omega t),$$

where $\boldsymbol{\lambda}$ is a third-rank tensor:

$$\begin{aligned}\boldsymbol{\lambda}&=(2\omega_{\mathbf{q}}/\hbar)\textstyle\sum_{n_{\mathbf{q}}}(n+1)_{\mathbf{q}}\boldsymbol{\xi}(\rho_{n_{\mathbf{q}}}{}^0-\rho_{(n+1)_{\mathbf{q}}}{}^0),\\ \boldsymbol{\xi}&=\{\mathbf{e}(\mathbf{q},b)\}(1/\hbar)\textstyle\sum_I(n+1)_{\mathbf{q}}{}^{-1/2}N^{1/2}\left[\dfrac{\langle(n+1)_{\mathbf{q}},g|\sum_b(e\mathbf{r})_{bS}|I\rangle\langle I|\sum_b(e\mathbf{r})_{bl}|n_{\mathbf{q}},g\rangle}{\omega_l-\omega_{I,gn_{\mathbf{q}}}}-\dfrac{\langle(n+1)_{\mathbf{q}},g|\sum_b(e\mathbf{r})_{bl}|I\rangle\langle I|\sum_b(e\mathbf{r})_{bS}|n_{\mathbf{q}},g\rangle}{\omega_S+\omega_{I,gn_{\mathbf{q}}}}\right]\exp(i\mathbf{q}\cdot\mathbf{R}_m).\end{aligned}\tag{11}$$

The square bracket in the expression for $\boldsymbol{\xi}$ is likely to be proportional to $(n+1)_{\mathbf{q}}{}^{1/2}$, since the states $\langle(n+1)_{\mathbf{q}}|$ and $|n_{\mathbf{q}}\rangle$ must be connected implicitly by the operator $U^+(q,b)$ or $a_{\mathbf{q}}$. The factor $N^{1/2}$ in $\boldsymbol{\xi}$ arises as a normalization factor attached to the states $\langle(n+1)_{\mathbf{q}}|$ and $|n_{\mathbf{q}}\rangle$ because of the definition of $U_\pm(q,b)$ in Eq. (5). The quantity $\boldsymbol{\xi}$, which has the dimension of an atomic polarizability, is then independent of $n_{\mathbf{q}}$, and since

$$\textstyle\sum_{n_{\mathbf{q}}}(n+1)_{\mathbf{q}}(\rho_{n_{\mathbf{q}}}{}^0-\rho_{(n+1)_{\mathbf{q}}}{}^0)=\sum_{n_{\mathbf{q}}}\rho_{n_{\mathbf{q}}}{}^0=1,$$

Eq. (11) gives

$$\boldsymbol{\lambda}=(2\omega_{\mathbf{q}}/\hbar)\boldsymbol{\xi}.\tag{11a}$$

This can be shown explicitly for the case where the electron-phonon interaction is small so that it can be treated as a small perturbation, mixing the states. The intermediate state can be written as $\langle I| = \langle (n+1)_{\mathbf{q}}, i|$, and

$$\begin{aligned}\langle (n+1)_{\mathbf{q}}, i|e\mathbf{r}|n_{\mathbf{q}},g\rangle &= \frac{\langle i|e\mathbf{r}|g\rangle}{h\omega_{\mathbf{q}}}[\langle (n+1)_{\mathbf{q}}, i|\mathcal{H}_{\text{e-p}}|n_{\mathbf{q}},i\rangle - \langle (n+1)_{\mathbf{q}}, g|\mathcal{H}_{\text{e-p}}|n_{\mathbf{q}},g\rangle],\\ \langle (n+1)_{\mathbf{q}}, g|e\mathbf{r}|n_{\mathbf{q}},i\rangle &= \frac{\langle g|e\mathbf{r}|i\rangle}{-\hbar\omega_{\mathbf{q}}}[\langle (n+1)_{\mathbf{q}}, i|\mathcal{H}_{\text{e-p}}|n_{\mathbf{q}},i\rangle - \langle (n+1)_{\mathbf{q}}, g|\mathcal{H}_{\text{e-p}}|n_{\mathbf{q}},g\rangle].\end{aligned} \tag{12}$$

With $\mathcal{H}_{\text{e-p}}$ given by Eq. (4), and substituting the above expression into Eq. (11), one finds

$$\begin{aligned}\boldsymbol{\xi} &= \{\mathbf{e}(\mathbf{q},b)\}(1/\hbar)[\langle g|\textstyle\sum_b(e\mathbf{r})_{bS}|i\rangle\langle i|\sum_b(e\mathbf{r})_{bl}|g\rangle(\langle i|\sum_b(\mathbf{f}\cdot\hat{e}^*)_b|i\rangle - \langle g|\sum_b(\mathbf{f}\cdot\hat{e}^*)_b|g\rangle)](A/\omega_{\mathbf{q}}), \\ A &= [(\omega_S-\omega_{ng})^{-1} - (\omega_l-\omega_{ng})^{-1} + (\omega_S+\omega_{ng})^{-1} - (\omega_l+\omega_{ng})^{-1}]/\hbar,\end{aligned} \tag{13}$$

where **f** is the generalized force in Eq. (4). The phonon wave is coupled to the laser and Stokes fields $\langle\mathbf{E}_l\rangle$ and $\langle\mathbf{E}_S\rangle$ to give rise to the stimulated Raman and Brillouin effects. The wave equation for $\langle\mathbf{E}_l\rangle$ and $\langle\mathbf{E}_S\rangle$ is

$$\nabla^2\langle\mathbf{E}_{l,S}\rangle + (\omega_{l,S}^2\varepsilon_{l,S}/c^2)\langle\mathbf{E}_{l,S}\rangle = -(4\pi\omega_{l,S}^2/c^2)\langle\mathbf{P}_{l,S}^{NL}\rangle, \tag{14}$$

where $\langle\mathbf{P}_{l,S}^{NL}\rangle$ is obtained from the usual iterative procedure in the density matrix formalism. In particular,

$$\langle\mathbf{P}_S^{NL}(\mathbf{R},t)\rangle = \mathfrak{N}\boldsymbol{\xi}^\dagger\boldsymbol{\varepsilon}_l\langle\mathbf{Q}_{\mathbf{q}}(g\mathbf{R},t)\rangle, \tag{15}$$

where $\mathfrak{N}$ is the number of unit cells per unit volume. The Boltzmann factors ρ_n^0 disappear in the nonlinear coupling terms of both Eq. (11) and Eq. (15). Therefore, the stimulated Raman gain, obtained from the solution of the coupled wave equations for $\langle\mathbf{E}_S\rangle$ and $\langle\mathbf{Q}_{\mathbf{q}}\rangle$ would be independent of the average thermal excitations of phonons.

For acoustic phonons the harmonic approximation on which the linear expression for the displacement operator U is based, is nearly always valid. The effect of anharmonic terms may be taken into account as a damping term, caused by collisions between the acoustic waves. The temperature dependence of the stimulated Brillouin effect is entirely contained in the temperature dependence of the damping constant Γ. Even though the concept of elementary excitations breaks down at high temperature, the classical acoustic wave can still be described in the same manner, even in liquids.

The situation is different for optical phonons. In this case the anharmonicity of the molecular vibrations limits the validity of the harmonic collective excitations to the low-temperature regime, where the probability to have an excitation at a particular localized site is small compared to unity.

The dispersion law for optical phonons is very different from that of acoustic phonons. The contribution of the collective motion to the wavelength-dependent part of the energy is small and the damping is relatively large, $\beta q^2 \ll \omega_{\mathbf{q}}\Gamma$. Under these circumstances it is appropriate to consider the localized vibrational excitations of individual molecules.[8] Since the vibrations are strongly anharmonic, only the ground state and the first vibrational level need be considered. It is a well-known result for this case of individual molecules that the Raman susceptibility is proportional to the population difference in these two states, $\rho_0^0-\rho_1^0$. This temperature dependence through the Boltzmann factors does not appear in the calculation with collective elementary excitation waves, which is strictly valid only at absolute zero. The case of optical phonons derived for a lattice array of molecules wlth two vibrational levels is analogous to the case of spin waves derived from a lattice of spins with $S=\frac{1}{2}$. The representation by elementary excitations with boson characteristics is a low-temperature approximation.

The formalism of the coupling of light with optical phonons may be taken over to the case of spin waves. The coupling of light with plasma waves has been discussed elsewhere.[13]

III. COUPLING OF LIGHT WITH MAGNONS

The electronic Hamiltonian for a magnetic system consists of spin and orbital parts. The spin part, with exchange interaction among spins, forms the magnon system. The radiation field is treated classically and is again assumed to consist of two waves, E_l and E_S. The total Hamiltonian is written as

$$\mathcal{H} = \mathcal{H}_{\text{magnon}} + \mathcal{H}_{\text{orb}} + \mathcal{H}_{\text{int}}. \tag{16}$$

The nuclear vibrational part is omitted in this section. The interaction Hamiltonian consists of spin-orbit, spin-radiation, and orbit-radiation interactions,

$$\mathcal{H}_{\text{int}} = \mathcal{H}_{\text{S-L}} + \mathcal{H}_{\text{L-r}} + \mathcal{H}_{\text{S-r}}. \tag{17}$$

These interactions have the familiar bilinear form

$$\begin{aligned}\mathcal{H}_{\text{S-L}} &= \sum_{m,b}\lambda_{mb}\mathbf{L}_{mb}\cdot\mathbf{S}_{mb},\\ \mathcal{H}_{\text{L-r}} &= -\sum_{m,b}[e\mathbf{r}_{mb}\cdot\mathbf{E}_{mb} + \mu\mathbf{L}_{mb}\cdot\mathbf{H}_{mb}],\\ \mathcal{H}_{\text{S-r}} &= -\sum_{m,b}2\mu\mathbf{S}_{mb}\cdot\mathbf{H}_{mb},\end{aligned} \tag{18}$$

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[13] N. Bloembergen and Y. R. Shen, Phys. Rev., **141**, 298 (1966).

where λ and μ are the spin-orbit coupling constant and the Bohr magneton, respectively. The electric and magnetic fields of the radiation at the bth atom in the mth unit cell are designated by E_{mb} and H_{mb}. The terms $\mu\mathbf{L}\cdot\mathbf{H}$ and $2\mu\mathbf{S}\cdot\mathbf{H}$ correspond to magnetic-dipole transitions. Javan and Winter first suggested the stimulated Raman maser action in a paramagnetic two-level spin system [compare Fig. 1(a)]. In the optical spin Raman transitions, shown in Figs. 1(b) and 1(c), the intermediate states can be connected by the electric-dipole interaction $e\mathbf{r}\cdot\mathbf{E}$. The magnetic-dipole terms $\mu\mathbf{L}\cdot\mathbf{H}$ and $2\mu\mathbf{S}\cdot\mathbf{H}$ are negligible in comparison. The interaction Hamiltonian reduces in these cases to a form similar to the one in the previous section. The role of $\mathcal{H}_{\text{e-p}}$ is taken over by $\mathcal{H}_{\text{S-L}}$.

The magnon Hamiltonian is

$$\mathcal{H}_{\text{magnon}}=-\sum_{\substack{m,m'\\b,b'}} J_{mb-mb'}\mathbf{S}_{mb}\cdot\mathbf{S}_{m'b'}-2\mu\mathbf{H}_0\cdot\sum_{m,b}\mathbf{S}_{mb}\,, \tag{19}$$

where J is the exchange coupling constant and $\mathbf{H}_0$ the dc magnetic field. In the harmonic approximation, $\mathcal{H}_{\text{magnon}}$ can be quantized as[14]

$$\mathcal{H}_{\text{magnon}}=\sum_{\mathbf{q},j}\hbar\omega_{\mathbf{q}j}(a_{\mathbf{q}j}{}^{\dagger}a_{\mathbf{q}j}+\tfrac{1}{2})\,. \tag{20}$$

The spin component is expressed in terms of creation and annihilation operators $a_{\mathbf{q}j}{}^{\dagger}$, $a_{\mathbf{q}j}$ from the linearized Holstein-Primakoff transformation[15]

$$\begin{aligned} S_{mb}{}^{+}=(S_{mb})_x+i(S_{mb})_y&=(2S_b/N)^{1/2}\sum_{\mathbf{q},j}e(\mathbf{q},b,j)a_{\mathbf{q}j}\exp(i\mathbf{q}\cdot\mathbf{R}_m)\\ &=\sum_{\mathbf{q},j}S(\mathbf{q},j)\exp(i\mathbf{q}\cdot\mathbf{R}_m)\,,\\ (S_{mb})^2=S_b(S_b+1)\,. \end{aligned} \tag{21}$$

The magnon modes $\omega_{\mathbf{q}j}$ are obtained by solving the set of linearized Bloch equations of motion for $S_{mb}{}^{+}$ for the magnetically inequivalent atoms in a unit cell,[16] just as in the case of phonons. The number of magnon branches is of course equal to the number of magnetically inequivalent atoms in each unit cell. In the long-wavelength limit,

$$\omega_{\mathbf{q}j}=\omega_0(\mathbf{H}_0,J)+\beta(J)q^2\,. \tag{22}$$

In particular, when there is only one magnetic sublattice with one magnetic atom per unit cell, there is only one magnon branch with $\omega_0=2\mu H_0$ and $\beta=JSa^2$, where a is the lattice constant.

The magnon wave equation can now be derived from the density matrix formalism. Let ρ be the density matrix operator for the material system. We shall again assume only two states for the orbital part of each atom, $\langle i|_{mb}$ and $\langle g|_{mb}$ and $\langle G|=\prod_{m,b}\langle g|_{mb}$. Consider the equation for $\langle(n+1)_{\mathbf{q}},G|\rho|n_{\mathbf{q}},G\rangle$, where $\langle n_{\mathbf{q}}|$ denotes the excitation of the magnon wavelength wave vector $\mathbf{q}=\mathbf{k}_l-\mathbf{k}_S$. From the equation of motion for ρ and Eqs. (16)–(20), we find

$$\left(i\frac{\partial}{\partial t}-\omega_{\mathbf{q}}+i\Gamma\right)\langle(n+1)_{\mathbf{q}},G|\rho^{(\omega)}|n_{\mathbf{q}},G\rangle e^{-i\omega t}=\frac{1}{\hbar}\langle(n+1)_{\mathbf{q}},G|[\mathcal{H}_{\text{e-r}},\rho]^{(\omega)}|n_{\mathbf{q}},G\rangle e^{-i\omega t}\,, \tag{23}$$

where $\omega=\omega_l-\omega_S$. The lowest order nonvanishing result in the perturbation expansion of $[\mathcal{H}_{\text{e-r}},\rho]^{(\omega)}$ gives

$$\begin{aligned}\langle(n+1)_{\mathbf{q}},G|[\mathcal{H},\rho]^{(\omega)}|n_{\mathbf{q}},G\rangle=\sum_{m,I}\Bigg[&\frac{\langle(n+1)_{\mathbf{q}},g|\sum_b(e\mathbf{r}\cdot\boldsymbol{\varepsilon}_S{}^{*})_b|I\rangle\langle I|\sum_b(e\mathbf{r}\cdot\boldsymbol{\varepsilon}_l)_b|n_{\mathbf{q}},g\rangle}{\hbar(\omega_l-\omega_{I,gn})}\\ &-\frac{\langle(n+1)_{\mathbf{q}},g|\sum_b(e\mathbf{r}\cdot\boldsymbol{\varepsilon}_l)_b|I\rangle\langle I|\sum_b(e\mathbf{r}\cdot\boldsymbol{\varepsilon}_S{}^{*})_b|n_{\mathbf{q}},g\rangle}{\hbar(\omega_S+\omega_{I,gn})}\Bigg](\rho_{n_{\mathbf{q}}}{}^{0}-\rho_{(n+1)_{\mathbf{q}}}{}^{0})\exp(i\mathbf{q}\cdot\mathbf{R}_m)\,.\end{aligned} \tag{24}$$

Here, the intermediate state with arbitrary mixing of spin and orbital character is denoted by $\langle I|$, and $\omega_{I,gn}$ is the frequency separation between $\langle I|$ and $\langle n_{\mathbf{q}},g|$. Since $\mathcal{H}_{\text{e-r}}$ cannot change the occupation number of magnons, Eq. (24) would vanish if the spin-orbit interaction $\mathcal{H}_{\text{S-L}}$ were not present. When $\mathcal{H}_{\text{S-L}}$ is small, it can be treated as a perturba-

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[14] See, for example, C. Kittel, *Quantum Theory of Solids* (John Wiley & Sons, Inc., New York, 1964).

[15] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

[16] See, for example, B. Harris, Phys. Rev. **132**, 2398 (1963).

tion in mixing the states. With the intermediate state written as $\langle I| = \langle n_{\mathbf{q}},i|$, one finds

$$\begin{aligned}\langle (n+1)_{\mathbf{q}},\, i|e\mathbf{r}|n_{\mathbf{q}},g\rangle &= \sum_{i'}\Bigg[\frac{\langle (n+1)_{\mathbf{q}},\, i|\mathcal{H}_{\text{S-L}}|n_{\mathbf{q}},i'\rangle\langle n_{\mathbf{q}},i'|e\mathbf{r}|n_{\mathbf{q}},g\rangle}{\hbar\omega_{i(n+1),\,i'n}} \\ &\qquad + \frac{\langle (n+1)_{\mathbf{q}},\, i|e\mathbf{r}|\,(n+1)_{\mathbf{q}}i'\rangle\langle (n+1)_{\mathbf{q}},\, i'|\mathcal{H}_{\text{S-L}}|n_{\mathbf{q}},g\rangle}{\hbar\omega_{gn,\,i'(n+1)}}\Bigg], \\ \langle (n+1)_{\mathbf{q}},\, g|e\mathbf{r}|n_{\mathbf{q}},i\rangle &= \sum_{i'}\Bigg[\frac{\langle (n+1)_{\mathbf{q}},\, g|e\mathbf{r}|\,(n+1)_{\mathbf{q}},\, i'\rangle\langle (n+1)_{\mathbf{q}},\, i'|\mathcal{H}_{\text{S-L}}|n_{\mathbf{q}},i\rangle}{\hbar\omega_{in,\,i'(n+1)}} \\ &\qquad + \frac{\langle (n+1)_{\mathbf{q}},\, g|\mathcal{H}_{\text{S-L}}|n_{\mathbf{q}},i'\rangle\langle n_{\mathbf{q}},i'|e\mathbf{r}|n_{\mathbf{q}},i\rangle}{\hbar\omega_{g(n+1),\,i'n}}\Bigg].\end{aligned} \tag{25}$$

These equations may apply for certain iron-group ions. If $\langle g|$ and $\langle i|$ denote eigenstates in the crystalline field potential, the quenching of the orbital angular momentum implies that the diagonal elements of the spin-orbit interaction vanish

$$\langle (n+1)_{\mathbf{q}},\, i|\mathcal{H}_{\text{S-L}}|n_{\mathbf{q}},i\rangle = \langle (n+1)_{\mathbf{q}},\, g|\mathcal{H}_{\text{S-L}}|n_{\mathbf{q}},g\rangle = 0.$$

This is different from the phonon case described by Eq. (12).

The spin wave is defined as

$$\langle S_{\mathbf{q}}(R,t)\rangle = \langle G|\operatorname{Tr}\textstyle\sum_b (1/2S_b)^{1/2}\rho S_b^{+}(\mathbf{q},\omega)|G\rangle \exp(i\mathbf{q}\cdot\mathbf{R}-i\omega t). \tag{26}$$

Equation (23) then leads to the equation for the spin wave

$$\begin{gathered}\left[i\frac{\partial}{\partial t}-\omega_0+i\Gamma+\beta\nabla^2\right]\langle S_{\mathbf{q}}(\mathbf{R},t)\rangle = \lambda_{\text{S}}:\boldsymbol{\varepsilon}_l\boldsymbol{\varepsilon}_S^{*}\exp(i\mathbf{q}\cdot\mathbf{R}-i\omega t), \\ \lambda_{\text{S}} = \hbar^{-1}\textstyle\sum_{n_{\mathbf{q}}}(n+1)_{\mathbf{q}}\xi_{\text{S}}(\rho_{n_{\mathbf{q}}}{}^{0}-\rho_{(n+1)_{\mathbf{q}}}{}^{0}), \\ \xi_{\text{S}} = (1/\hbar)\{e(\mathbf{q},b)\}\textstyle\sum_I N^{1/2}(n+1)_{\mathbf{q}}{}^{-1/2}\Bigg[\dfrac{\langle (n+1)_{\mathbf{q}},\, g|\sum_b(e\mathbf{r})_{bS}|I\rangle\langle I|\sum_b(e\mathbf{r})_{bl}|n_{\mathbf{q}},g\rangle}{\omega_l-\omega_{I,gn}} \\ -\dfrac{\langle (n+1)_{\mathbf{q}},\, g|\sum_b(e\mathbf{r})_{bl}|I\rangle\langle I|\sum_b(e\mathbf{r})_{bS}|n_{\mathbf{q}},g\rangle}{\omega_S+\omega_{I,gn}}\Bigg]\exp(i\mathbf{q}\cdot\mathbf{R}_m).\end{gathered} \tag{27}$$

Note the similarity of this expression and the corresponding Eq. (11) for the phonon case. The expression in square brackets can be evaluated explicitly in the case of weak spin-orbit coupling with Eq. (25). It is seen to be proportional to $(n+1)_{\mathbf{q}}{}^{1/2}$. This result has probably more general validity, as the term in square brackets connects two boson eigenstates differing by one unit of excitation. The factor $N^{1/2}$ in the expression again arises as a normalization factor attached to the states $\langle (n+1)_{\mathbf{q}}|$ and $|n_{\mathbf{q}}\rangle$. The expression for λ_{S} is independent of the $\rho_{n_{\mathbf{q}}}{}^{0}$ in this case by the same arguments which led to Eq. (11a) and, in fact $\lambda_{\text{S}} = \xi_{\text{S}}/\hbar$. The coupling constant is therefore independent of temperature in this harmonic approximation. This result can only be expected to have validity for temperatures well below the Curie or Néel temperature. When T becomes an appreciable fraction of T_c higher order terms in the spin-wave variables can no longer be ignored in the Holstein-Primakoff transformation. This is the usual restriction on the validity of spin-wave theories.

The spin wave is coupled to the two light waves $\mathbf{E}_l$ and $\mathbf{E}_S$ to give rise to the spin-Raman effect. The wave equation for the Stokes wave is

$$\begin{aligned}\nabla^2\mathbf{E}_S(\mathbf{R},t)+(\omega_S{}^2\boldsymbol{\varepsilon}_S/c^2)\mathbf{E}_S(\mathbf{R},t) \\ = -(4\pi\omega_S{}^2/c^2)\langle\mathbf{P}_S{}^{NL}(\mathbf{R},t)\rangle.\end{aligned} \tag{28}$$

The nonlinear polarization in Eq. (28) can be found by the usual perturbation calculation,

$$\mathbf{P}_S{}^{NL}(R,t) = \mathfrak{N}\xi_{\text{S}}{}^{\dagger}\boldsymbol{\varepsilon}_l\langle S(\mathbf{q},\omega)\rangle^{*}\exp(i\mathbf{q}\cdot\mathbf{R}-i\omega t), \tag{29}$$

where $\mathfrak{N}$ is the number of unit spin cells per unit volume.

The above derivation is very similar to that in the coupled photon-phonon case. If the spin-orbit coupling and the crystalline-field interaction are large, a pure spin wave of course does not exist. The formal derivation remains valid in this case, which is represented by Fig. 1(c).

The gain coefficient for the stimulated spin Raman effect[6,8] can now be solved from the set of coupled wave equations (27) and (28) with λ_{S} replaced by ξ_{S}. For infinite plane waves with linear polarization in a medium of plane boundaries, the Stokes wave vector is found to be

$$\mathbf{k}_{S\pm}=\mathbf{k}_S{}^0+\hat{z}\left\{\frac{1}{2}(i\alpha_S-D/2\beta q_z{}^0)\pm\frac{1}{2}\left[\left(\frac{D^*}{2\beta q_z{}^0}+i\alpha_S\right)^2-\frac{2\mathfrak{N}\pi\omega_S{}^2\xi_S{}^2|\mathcal{E}_l|^2}{c^2\beta k_{Sz}{}^0q_z{}^0\hbar}\right]^{1/2}\right\}, \quad (30)$$

where

$$\begin{aligned} k_S{}^0&=\omega_S{}^2\epsilon_S'/c^2, \\ \alpha_S&=\omega_S{}^2\epsilon_S''/2c^2k_{Sz}{}^0, \\ D^*&=\omega-\omega_0-\beta q^{0^2}-i\Gamma_S, \\ \mathbf{q}^0&=\mathbf{k}_l-\mathbf{k}_S{}^0, \\ \mathbf{q}'&=\mathbf{k}_l'-\mathbf{k}_S' \quad \text{and} \quad k_S''=q''. \end{aligned} \quad (31)$$

The laser field $\mathcal{E}_l$ is assumed to be a constant parameter and the unit vector $\hat{z}$ is normal to the boundaries of the Raman cell. The imaginary part of k_S is the gain or loss coefficient. The corresponding waves are given by

$$E_S=[C_{S+}\exp(i\mathbf{k}_{S+}\cdot\mathbf{r})+C_{S-}\exp(i\mathbf{k}_{S-}\cdot\mathbf{r})]e^{-i\omega_St},$$
$$\langle S\rangle=[C_{S+}'\exp(i\mathbf{q}_+\cdot\mathbf{r})+C_{S-}'\exp(i\mathbf{q}_-\cdot\mathbf{r})]e^{-i\omega t},$$
$$C_{S\pm}/C_{S\pm}'=\mathfrak{N}(\pi\omega_S{}^2/c^2k_{Sz}{}^0)\xi_S\mathcal{E}_l/(k_S-k_S{}^0+i\alpha).$$

If the spin wave is highly damped, such that

$$(D^*/2\beta q_z{}^0+i\alpha_S)^2\gg 2\mathfrak{N}\pi\omega_S{}^2\xi_S{}^2|\mathcal{E}_l|^2/c^2\beta q_z{}^0k_{Sz}{}^0\hbar,$$

the square root in Eq. (30) can be expanded into a power series to give a gain coefficient

$$\mathrm{Im}k_{S+}=\alpha_S+\mathrm{Im}\left\{\frac{2\mathfrak{N}\pi\omega_S{}^2\xi_S{}^2|\mathcal{E}_l|^2}{c^2k_{Sz}{}^0D^*\hbar}\right\}. \quad (32)$$

The gain is a maximum when both linear momentum and energy matching conditions are satisfied, i.e.,

$$\mathbf{k}_l=\mathbf{k}_S{}^0+\mathbf{q}^0 \quad \text{and} \quad \omega_l=\omega_S+\omega.$$

This is indicated by the resonance point R in Fig. 2. Curves 2 and 3 in this figure are given by

$$q^0=|\mathbf{k}_l-\mathbf{k}_S{}^0|=[\omega_l(n_l\hat{k}_l-n_S\hat{k}_S{}^0)\cdot\hat{q}^0+\omega_Sn_S\hat{k}_S{}^0\cdot\hat{q}^0]/c$$

for the forward and backward scattering, respectively. Here, n's are the indices of refraction, and $\hat{k}$'s the unit vectors. The Raman susceptibility corresponding to the Raman gain of Eq. (32) assumes the very simple form,[8]

$$\chi_{\text{Raman}}{}^{\text{spin}}=\mathfrak{N}\xi_S{}^2/\hbar\Gamma_S, \quad (33)$$

where ξ_S is given by Eq. (27) and has the dimension of an atomic polarizability, and Γ_S is the damping constant for the spin wave.

When the probability for spin excitation at a localized site is not very small compared to unity, this harmonic approximation of the spin waves loses its validity. In the opposite limit of very high temperature, above the Curie or Néel point, it is clearly more appropriate to consider the energy levels of localized spins. In this paramagnetic case, the Boltzmann factor $\rho_0{}^0-\rho_1{}^0$ appears. The effect is proportional to the difference in population of the two magnetic levels concerned. If these levels form a Kramer's doublet, the temperature dependence is the same as the paramagnetic magnetization arising from these two levels. This suggests that the temperature dependence of the spin Raman transitions in a ferromagnet is similar to, although not necessarily identical to, the temperature dependence of the magnetization $M(T)$.

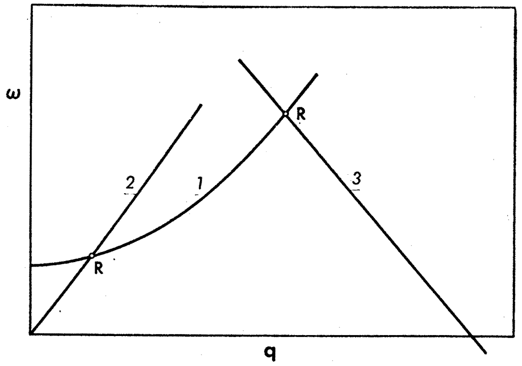

FIG. 2. Dispersion curves describing the spin Raman effect. Curve 1 is the dispersion curve for an acoustic magnon wave. Curves 2 and 3 describe the linear-momentum and energy-matching condition given by

$$q^0=|\mathbf{k}_l-\mathbf{k}_S{}^0|=[\omega_l(n_l\hat{k}_l-n_S\hat{k}_S{}^0)\cdot q^0+\omega n_S\hat{k}_S{}^0\cdot\hat{q}^0]/c$$

for the forward and backward Stokes scattering, respectively. The resonant points are denoted by R.

IV. THE SPIN RAMAN EFFECT IN PARA-, FERRO-, FERRI-, AND ANTIFERROMAGNETIC MATERIALS

The possibility of observing the Raman effect in magnetic systems was described in the introduction. Hough and Singh[4] observed the spontaneous Raman transitions between two electronic levels of Pr^{3+} in a LaF_3 crystal. The Raman scattering due to spin excitations, however, has not yet been observed.

Both the spontaneous and the stimulated Raman scattering depend on the coupling constant ξ_S. The Raman transition probability increases as $|\xi_S|$ increases. As shown in Eq. (27), the magnitude of ξ_S becomes large if (1) the frequency ω_l or ω_S (or both) approaches a resonance, and (2) the matrix elements are large. In some simple cases, there are also selection rules governing the Raman transitions.

Consider first a paramagnetic system with a small crystalline field, so that m is still a good magnetic quantum number. The localized spin model applies to this case. The Raman transition probability is proportional to $\mathfrak{N}|\xi_S|^2|\mathcal{E}_l|^2(\rho_0{}^0-\rho_1{}^0)/\Gamma$, where ξ_S is given by Eq. (27) with the magnon states replaced by the local paramagnetic states. The degeneracy of the magnetic states is of course lifted by the applied dc magnetic field. In order to have the initial and the final magnetic states, $\langle g|$ and $\langle f|$, in the normal Raman transitions connected by the electric-dipole operator, the magnetic quantum number must change by $\Delta m = m_f - m_i = 1$ or 2. With $\Delta m = 1$ [see Fig. 1(b)] the selection rule requires the laser polarization to have a circular component around, and the Stokes polarization to have a linear component along, the magnetic field, or vice versa. The exciting beam propagating along the magnetic field cannot excite Stokes scattering in the forward direction. In the spontaneous Raman emission, one observes preferably at right angles to the incoming beam. One would find a linearly polarized Stokes light if the exciting beam is propagating in the direction of the magnetization, and a circularly polarized Stokes light if the exciting beam is propagating in the direction perpendicular to the magnetization. In the stimulated Raman effect, it is best to have the laser and the Stokes parallel to each other. They may then propagate at 45° with respect to the magnetic field. The geometry used by Dennis and Tennanwald[17] with the laser beam at an angle to the Stokes beam would also be possible. With $\Delta m = 2$, the selection rules require the polarizations of the laser and the Stokes to have components circulating in the same sense around the magnetic field. Again, the polarization properties depend in an interesting way on the direction of the magnetization and on the directions of propagation of the two beams. If the crystalline field is so large that m is no longer a good quantum number, the above selection rules in general break down.

Consider next a simple ferromagnet with a single sublattice, and assume the spin waves originating from individual ion states with pure magnetic quantum numbers $m = \pm\frac{1}{2}$. Only the acoustic magnon branch exists. The states corresponding to zero- and one-magnon excitation can be written as

$$\begin{aligned} \langle 0_{\mathbf{q}}| &= \prod_i \langle +|_i\,, \\ \langle 1_{\mathbf{q}}| &= (1/N)^{1/2} \sum_j \Big(\langle -|_j \prod_{i\neq j} \langle +|_i\Big) \exp(i\mathbf{q}\cdot\mathbf{R}_j)\,, \end{aligned} \tag{34}$$

where $\langle +|_i$ and $\langle -|_i$ are the two spin states for the ith spin. Substituting Eq. (34) into Eq. (27), one finds that ξ_S is nonvanishing only if the product of matrix elements of the type $\langle g,-|e\mathbf{r}|I\rangle\langle I|e\mathbf{r}|g,+\rangle$ is different from zero. This requires that the magnetic quantum numbers of the initial and the final states in the Raman transitions differ by $\Delta m = 1$. The selection rule governing the polarization properties for the laser and the Stokes discussed previously for the paramagnetic case again applies. If the crystalline field is large, the spin wave is no longer composed of pure spin states. Then, in general, the selection rule breaks down.

In ferromagnets with more than one sublattice, and in ferri- and antiferromagnets, the sublattices are coupled together through exchange coupling to give different magnon branches. The eigenmodes and the eigenvectors are obtained from the coupled Bloch equations for the magnetizations of the sublattices. Assume a ferrite with two sublattices A and B, the corresponding spins being S_A and S_B, respectively, with pure spin states $\langle \pm\frac{1}{2}|$ for each spin. Since the spins are pointing in opposite directions, we write $S_{Az} = S_a$ and $S_{Bz} = -S_b$, and $S_A^{\pm}|\pm\rangle_A = |\pm\rangle_A$ and $S_B^{\pm}|\pm\rangle_B = |\mp\rangle_B$. Also assume that the exchange coupling exists only between spins on different sublattices. The coupled Bloch equations for $S_A^{\pm}$ and $S_B^{\pm}$ yield the magnon exchange eigenfrequency, or optical magnon mode at $\mathbf{q} = 0$[18]

$$\begin{aligned} \omega_0 = \tfrac{1}{2}(\omega_{eB} + \omega_{aA} - \omega_{eA} - \omega_{aB}) + \tfrac{1}{2}[(\omega_{eA} + \omega_{eB} \\ + \omega_{aA} + \omega_{aB})^2 - 4\omega_{eA}\omega_{eB}]^{1/2}\,, \\ (\omega_{eB} + \omega_{eA}) \geqq (\omega_{eA} + \omega_{aB})\,, \end{aligned} \tag{35}$$

where ω_{eA}, ω_{eB}, ω_{aA}, and ω_{aB} are the exchange and anisotropic frequencies for the two spins, respectively. The corresponding eigenmode is

$$\begin{aligned} \langle S_A^{+}\rangle/\langle S_B^{+}\rangle &= -\alpha_1/\alpha_2\,, \\ \alpha_1 &= \omega_{eA}/D\,, \\ \alpha_2 &= (\omega_{eB} + \omega_{eA} - \omega_0)/D\,, \\ D &= [\omega_{eA}^2 + (\omega_{eB} + \omega_{aA} - \omega_0)^2]^{1/2}\,. \end{aligned} \tag{36}$$

The states with zero- and one-magnon excitation can be written as

$$\begin{aligned} \langle 0_{\mathbf{q}}| &= \textstyle\prod_i \langle -,+|_i\,, \\ \langle 1_{\mathbf{q}}| &= (1/N)^{1/2} \textstyle\sum_j [(\alpha_1\langle +,+|_j - \alpha_2\langle -,-|_j) \\ &\qquad \times \prod_{i\neq j} \langle -,+|_i] \exp(i\mathbf{q}\cdot R_j)\,, \end{aligned} \tag{37}$$

where $\langle \pm,\pm|_j$ are the combined spin states for the spins in the jth unit cell. We have $S_{Aj}|-+\rangle_j = |++\rangle_j$ and $S_{Bj}|-+\rangle_j = |--\rangle_j$. A typical term in Eq. (27) which contributes to the value of ξ_S is

$$\begin{aligned} &\alpha_1\langle g,+,+|(er_A^{+} + er_B^{+})|I\rangle\langle I|(er_A^{z} + er_B^{z})|g,-,+\rangle \\ &\quad - \alpha_2\langle g,-,-|(er_A^{+} + er_B^{+})|I\rangle \\ &\quad\quad \times \langle I|(er_A^{z} + er_B^{z})|g,-,+\rangle\,, \end{aligned} \tag{38}$$

or with the superscripts + and Z interchanged. This again requires that the polarization of one beam, the laser or the Stokes, has a circular component around and the polarization of the other beam has a linear component along the direction of magnetization. The situation here is very similar to that in the direct infrared excitation discussed by Tinkham for the case of rare-earth garnets.[19] There, the absorption coefficient is

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[17] J. H. Dennis and P. E. Tannenwald, Appl. Phys. Letters **5**, 58 (1964).

[18] T. Nagamiya, K. Yosida, and R. Kubo, Advan. Phys. **4**, 14 (1955).

[19] M. Tinkham, Phys. Rev. **124**, 311 (1961); A. J. Sievers and M. Tinkham, *ibid.* **124**, 321 (1961).

proportional to the square of the matrix elements

$$[\alpha_1\langle g,+,+|(M_A{}^+ + M_B{}^+)|g,-,+\rangle \\ -\alpha_2\langle g,-,-|(M_A{}^+ + M_B{}^+)|g,-,+\rangle].$$

Because of the antiparallel exchange coupling between the two sublattices, the two terms tend to cancel each other, so that the infrared absorption coefficient (and the Raman transition probability) becomes smaller when the exchange coupling increases relative to the anisotropy energy. The above analysis also applies to the case of antiferromagnets if one puts $S_a = S_b$, $\omega_{aA} = \omega_{bB}$, and $\omega_{eA} = \omega_{eB}$. The direct far-infrared excitation of magnons in the antiferromagnet FeF_2 has also been investigated by Tinkham.[20] The eigenfrequencies and the eigenmodes given in Eqs. (35) and (36) should be slightly modified in the presence of a dc applied magnetic field. If the crystalline field is large, the selection rule governing the polarizations of the beams again breaks down.

The magnitude of the spin Raman effect may be estimated as follows. Comparison of Eqs. (15) and (27) shows that the two coupling constants ξ and ξ_S are comparable in magnitude. In the case of optical phonons, ξ would be zero if the electron-phonon interaction were absent, it therefore suffers a reduction factor $\mathcal{H}_{\text{e-p}}$/(vibrational energy). In the case of magnons, ξ_S would be zero if there were no spin-orbit interaction; the reduction factor is $\mathcal{H}_{LS}$/(crystal field). The two coupling constants would be of the same order of magnitude if the matrix elements involved were the same. In practice, the ultraviolet oscillator strength for organic molecules is close to 1, but for magnetic ions it is usually less than 0.1.[21] If the reduction factors and the damping constants for the two cases are approximately the same, the spin Raman effect would be about 2 orders of magnitude smaller than the ordinary Raman process in liquids. The linewidth of the spin excitation at low temperatures seem to be comparable to the optical phonon linewidth which is about 1 cm^{-1}. For example, the antiferromagnetic resonance in FeF_2 has a width of 0.1 cm^{-1} at 1°K which increases with temperature as T^4.[20] The damping constant for the ferromagnetic spin excitations at room temperature, lies in the range $\Gamma_S \sim 10^9 - 10^{11}$ sec^{-1}. The narrowest ferromagnetic linewidth in a garnet is about 0.5 G or $\Gamma_S \sim 10^7$ sec^{-1} at low temperatures.[19]

An alternative way to estimate the order of magnitude of the spin Raman effect is by comparison with the optical rotatory power of the magnetic system.[22,23] Physically, the spin Raman effect and the Faraday rotation are closely related magneto-optical effects. The former is derivable from a thermodynamical potential connected with the coupling between light waves and a spin wave. The coupling energy per ion is $\xi_S E_l E_S{}^*\langle S(\omega-\omega_S)\rangle^*$. The Faraday rotation is derivable from a potential that gives the difference in coupling energy of a right-circular- and a left-circular-polarized light wave with a longitudinal dc magnetization. This time-averaged energy per ion is $\xi_{\mathrm{Far}}\{|E_l{}^+|^2 - |E_l{}^-|^2\} \times\langle S(0)\rangle$, where $2\xi_{\mathrm{Far}}$ is the difference of the right and the left circular polarizabilities. Both effects would vanish in the absence of spin-orbit coupling, and the coupling constants in the two cases are quite similar. The constant ξ_S has the same order of magnitude as the circular polarizability, and so has ξ_{Far}, if the two circular polarizabilities do not accidentally cancel each other. This is the case of some iron-group ions, such as $Mn^{2\dagger}$, etc. Such a relationship was also noted by Pershan and coworkers.[24] They were only concerned with light polarizations perpendicular to the magnetization. Thus, only $\Delta m = 0$ or $\Delta m = \pm 2$ Raman transitions occur in their geometry. For the excitation of magnetic spin waves, the $\Delta m = \pm 1$ transitions are significant. They require the presence of a light component parallel to the magnetization. The ratio ξ_{Far}/ξ_S depends of course on the detailed geometry, crystal field splitting and mixing of the magnetic states.

The rotary power at magnetic saturation is related to the Faraday susceptibility by[25]

$$\phi = 4\pi\mathfrak{N}\xi_{\mathrm{Fal}}(\omega/nc)\ \text{rad/cm},$$

where $\mathfrak{N}$ is the number of magnetic ions per *cc* and n is the index of refraction. For Eu^{2+} the rotary power per ion has been determined experimentally.[25] For light at the ruby wavelength one finds $\xi_{\mathrm{Far}} = 5\times10^{-27}$ esu.

If we take $\mathfrak{N} = 5\times10^{22}$ in Eq. (33) and $\Gamma_S \approx 10^{11}$ sec^{-1} for a typical ferromagnet, one finds $\chi_{\mathrm{Raman}}{}^{\mathrm{spin}} \approx 10^{-14}$ esu. This is about two orders of magnitude smaller than the Raman susceptibility of several liquids in which stimulated Raman emission has been observed.

In principle, all magnon branches can be excited through the Raman process. In a spin Raman laser, however, the mode with the highest gain would be dominant. The stimulated Raman process would also have to compete with the ordinary stimulated Raman and Brillouin scattering.

For a single magnetic lattice, the acoustic ferromagnetic spin wave is of course the only magnon mode. It is interesting to compare the Raman excitation of this magnon mode with the Brillouin scattering. In both cases, the dispersion of the mode frequency is quite strong so that the Stokes radiation in different directions has different frequencies. The Stokes radiation in the spin Raman scattering can also go in the forward direction. If a strong dc magnetic field is applied, the mode frequency for $q=0$ is still different from zero. The frequency of the spin waves in the forward direction would

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[20] R. C. Ohlmann and M. Tinkham, Phys. Rev. **123**, 425 (1961).

[21] Estimated from optical absorption data in rare-earth and iron-group ions. See, for example, B. R. Judd, Phys. Rev. **127**, 750 (1962).

[22] Y. R. Shen, Phys. Rev. **133**, A511 (1964).

[23] A. M. Clogston, J. Phys. Radium **20**, 151 (1959).

[24] J. P. van der Ziel, P. S. Pershan, and L. D. Malmstrom, Phys. Rev. Letters **15**, 190 (1965).

[25] Y. R. Shen and N. Bloembergen, Phys. Rev. **133**, A515 (1964).

be low in small external fields. The momentum-matching condition becomes unimportant if the length of the sample l is so small that $ql<1$, as the uncertainty in the wave vector $\sim 1/l$ exceeds the wave vector q itself.

Since stimulated Brillouin scattering has a threshold which is comparable to that of the ordinary Raman effect in liquids, it will often dominate the spin Raman effect. The dispersion law is more favorable for spin waves in the forward direction. Note that the frequency of the magnon mode can also be tuned by the applied dc magnetic field.

In using giant-pulse lasers, one may still use the steady-state solution of the coupled wave equations, if the group velocity (or $\beta^{1/2}$) is small. This is the case for ordinary Raman effect and should also be valid for optical spin waves (or exchange modes) and for acoustic spin waves in very high fields. Otherwise, the transient solution of the type developed by Kroll[26] for the Brillouin case must be used.

The more interesting aspect of the spin Raman effect lies in the optical magnon branches. This includes the antiferromagnetic spin waves. Polarization properties of the beams should be investigated to see whether the simple selection rules break down or not. Experiments on FeF_2 with a Stokes shift of 55 cm^{-1} would be quite interesting. The large Stokes shift makes optical detection relatively easy. Various garnets are also suitable for investigation, especially yttrium iron garnet which is quite transparent in the near infrared.

In paramagnetic materials, the spin Raman effect arises from isolated ions. The Raman effect between two magnetic sublevels of the ground state and the Raman effect between two electronic levels have thesa me nature. The latter process was found by Hougen and Singh[4] in $LaF_3{:}Pr^{3+}$. Their experimental results give assurance that the spin Raman process and the Brillouin process might have the same order of magnitude. Although the concentration of magnetic ions in paramagnetic salts would be low, the linewidth could be as narrow as 10^{-2} or even 10^{-4} cm^{-1}, corresponding to a few gauss. The gain is proportional to the average population difference between the magnetic sublevels. If only these two sublevels are populated, the paramagnetic spin Raman gain would be proportional to the magnetization. This effect could be observed in the forward direction and could be tuned by the dc magnetic field, as distinguished from the Brillouin effect.

It appears worthwhile to search experimentally both for the spontaneous and the stimulated spin Raman effect. For the former, a gas laser focused into crystals at low temperature would be appropriate to observe a scattered radiation with a small Stokes shift.[27] For the latter, a resonant cavity with a different feedback factor for spin Stokes and Brillouin-shifted radiation would be useful.

V. COUPLING OF LIGHT WITH PHONONS AND MAGNONS

In discussing the spin Raman effect, we have neglected the term $\mu(\mathbf{L}+2\mathbf{S})\cdot\mathbf{H}$ in the Hamiltonian. This term would add to each electric-dipole matrix element a magnetic-dipole counterpart. In addition, if the em mode at the magnon frequency is present, there is a direct coupling between this em wave and the spin wave. It is this direct coupling that gives rise to magnetic resonance and far-infrared magnon excitation. We have also neglected the nuclear motion which is responsible for the phonon waves.

In principle, all waves existing in the medium can be coupled together either linearly or nonlinearly. The coupling is, however, effective only when both linear momentum and energy matching conditions are satisfied. Consider the case where five waves are present in the medium, the laser and the Stokes waves at ω_l and ω_S, and the infrared (or micro-) wave, the acoustic magnon wave, and the acoustic phonon wave at ω with $\omega=\omega_l-\omega_S$. The laser and the Stokes waves can be coupled to the phonon and the magnon waves through the nonlinear Raman-type coupling, and to the infrared (or micro-) wave through a nonlinear susceptibility of mixed electric and magnetic dipole character. This coupling constant is given by λ_2 or λ_4 in Eqs. (39) and (40). The magnon wave can be coupled linearly to the phonon wave through the magnetoelastic coupling,[28] and to the infrared (or micro-) wave magnetic field through the magnetic-dipole interaction. The coupling constants λ_6 and λ_9 between the acoustic phonon and the infrared (or micro-) wave is negligibly small since the waves cannot be matched simultaneously in energy and momentum. If the laser field is treated as a constant parameter, the remaining four waves are linearly coupled. The coupled wave equations can be written as

$$
\begin{aligned}
&\nabla^2\mathbf{E}_S+(\omega_S{}^2\boldsymbol{\varepsilon}_S/c^2)\mathbf{E}_S=\lambda_1\mathbf{E}_l\langle S\rangle^*+\lambda_2\mathbf{E}_l\mathbf{E}_v{}^*+\lambda_3\mathbf{E}_l\mathbf{A}_v{}^*,\\
&\nabla^2\mathbf{E}_v{}^*+(\omega^2\boldsymbol{\varepsilon}_v{}^*/c^2)\mathbf{E}_v{}^*=\lambda_4\mathbf{E}_l{}^*\mathbf{E}_S+\lambda_5\langle S\rangle^*+\lambda_6\mathbf{A}_v{}^*,\\
&\nabla^2\mathbf{A}_v{}^*+(\rho_a\omega^2/\mathbf{C}_a)\mathbf{A}_v{}^*-i(\rho_a/\mathbf{C}_a)2\omega\Gamma\mathbf{A}_v{}^*\\
&\qquad\qquad=\lambda_7\mathbf{E}_l{}^*\mathbf{E}_S+\lambda_8\langle S\rangle^*+\lambda_9\mathbf{E}_v{}^*,\\
&\nabla^2\langle S\rangle^*+(1/\beta)(\omega-\omega_0-i\Gamma_{\mathrm{S}})\langle S\rangle^*\\
&\qquad\qquad=\lambda_{10}\mathbf{E}_v{}^*+\lambda_{11}\mathbf{A}_v{}^*+\lambda_{12}\mathbf{E}_l{}^*\mathbf{E}_S,
\end{aligned}
\tag{39}
$$

where $\mathbf{A}_v$ denotes the acoustic vibrational wave. The coupling constants are either related to physical constants or can be derived explicitly.

$$
\begin{aligned}
&\lambda_1=-\mathfrak{N}(4\pi\omega_S{}^2/c^2)\xi_{\mathrm{s}}{}^\dagger,\\
&\lambda_2\sim(4\pi\omega_S{}^2/c^2)\\
&\qquad\times\mathfrak{N}\frac{(e\mathbf{r})_{gn}(e\mathbf{r})_{ng'}[\mu\epsilon^{1/2}(\mathbf{L}+2\mathbf{S})\times\hat{q}]_{gg'}}{\hbar^2(\omega_{l,S}-\omega_{ng})(\omega+\omega_{g'g})}.
\end{aligned}
\tag{40a}
$$

This coupling between three em waves in a medium with inversion symmetry is about 4 to 6 orders of

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[26] N. Kroll, J. Appl. Phys. **36**, 34 (1965).

[27] R. Y. Chiao and B. P. Stoicheff, J. Opt. Soc. Am. **54**, 1286 (1964); T. C. Daman, R. C. C. Leits, and S. P. S. Porto, Phys. Rev. Letters **14**, 9 (1965).

[28] C. Kittel, Phys. Rev. **110**, 836 (1958).

magnitude smaller than the coupling parameter for the second-harmonic generation in piezoelectric crystals. The magnetic-dipole term makes this term negligibly small compared to the other types of coupling.

$$\lambda_3 \sim (4\pi\omega_S^2/c^2)(-i\mathbf{q}\cdot\mathbf{p}). \tag{40b}$$

$\mathbf{p}$ is the photoelastic tensor, whose elements are of order unity

$$\begin{aligned} &\lambda_4 = \lambda_2, \\ &\lambda_5 = \mathfrak{N}[\mu\epsilon^{1/2}(\mathbf{L}+2\mathbf{S})\times\hat{q}]_{gg'}\sqrt{S_b}/S_{gg}^{+}. \end{aligned} \tag{40c}$$

This is the magnetic resonance term, that couples the magnetic field at the resonance frequency to the spin wave.

$$\begin{aligned} &\lambda_6 \sim 0, \\ &\lambda_7 \sim (i\mathbf{q}\cdot\mathbf{p})/\mathbf{C}_a^{-1}. \end{aligned} \tag{40d}$$

Here p is the photoelastic tensor and $\mathbf{C}_a$ is the elastic modulus tensor;

$$\lambda_8 = i\mathbf{q}b_2/\mathbf{C}_a. \tag{40e}$$

This magnetoelastic coupling is derived from the interaction Hamiltonian

$$\mathcal{H}_{me} = 2b_2[(S_x/S)\sigma_{zx} + (S_y/S)\sigma_{yz}],$$

where

$$\sigma_{yz} = (1/2)(\partial A_y/\partial z + \partial A_z/\partial y)$$

and

$$\sigma_{zx} = (1/2)(\partial A_z/\partial x + \partial A_x/\partial z)$$

are the shear strain components. The static magnetization is along the z direction. This is the only magnetoelastic coupling term in a cubic crystal which is linear in the spin variable. For a normal ferromagnet, the magnetoelastic constant b_2 has the magnitude of the order of 5×10^{-7} erg/cm^3.[28]

$$\begin{aligned} &\lambda_9 \sim 0, \\ &\lambda_{10} = -\frac{1}{\hbar}[\mu\epsilon^{1/2}(\mathbf{L}+2\mathbf{S})\times\hat{q}]_{g'g}S_{gg'}^{+}/\beta, \\ &\lambda_{11} = i\mathbf{q}b_2S_0\mathfrak{v}/\hbar\mu. \end{aligned} \tag{40f}$$

In this elastomagnetic coupling constant $\mathfrak{v}$ is the volume of a unit cell.

$$\lambda_{12} = -\xi_S/\hbar\beta. \tag{40g}$$

λ_1 and λ_{12} are the spin-light coupling constants described in Sec. III. Here, with $E_l = \mathcal{E}_l \exp[i\mathbf{k}_l\cdot\mathbf{r} - i\omega_l t]$, a solution of the set of coupled equations (39) takes the form

$$\begin{aligned} &E_S \sim \exp[i\mathbf{k}_S\cdot\mathbf{r} - i\omega_S t], \\ &E_v, A_v, \langle S\rangle \sim \exp[i\mathbf{q}\cdot\mathbf{r} - i\omega t]. \end{aligned} \tag{41}$$

The complex wave vectors $\mathbf{k}_S$ and $\mathbf{q} = \mathbf{k}_l - \mathbf{k}_S^*$ are obtained from the determinant

$$\begin{vmatrix} -(k_S^2 - \omega_S^2\epsilon_S/c^2) & -\lambda_1\mathcal{E}_l & -\lambda_2\mathcal{E}_l & -\lambda_3\mathcal{E}_l \\ -\lambda_{12}\mathcal{E}_l^* & -q^2 + (\omega - \omega_0 - i\Gamma_S)/\beta & -\lambda_{10} & -\lambda_{11} \\ -\lambda_4\mathcal{E}_l^* & -\lambda_5 & -q^2 + \omega_v^2\epsilon_v^*/c^2 & 0 \\ -\lambda_7\mathcal{E}_l^* & -\lambda_8 & 0 & -q^2 + (\rho_a/c_a)(\omega^2 - i2\omega\Gamma) \end{vmatrix} = 0. \tag{42}$$

The imaginary part of k_S gives the Stokes gain.

It is quite difficult to find the solution of Eq. (42). However, the dispersion relations of the waves are such that in general only three waves can be effectively coupled, since the infrared wave E_v is always decoupled from the acoustic wave A_v.

Consider first the coupling of E_S, A_v, and $\langle S\rangle$. The problem is essentially the same as the problem of the Raman effect in a polar medium[11] where the Stokes wave is coupled to the infrared and the optical phonon waves. That calculation can be carried over to the present case. Let

$$\begin{aligned} &\mathbf{k}_S = \mathbf{k}_S^0 + (\Delta K)\hat{z}, && (k_S^0)^2 = \omega_S\epsilon_S'/c^2, \\ &\mathbf{k}_l' = \mathbf{k}_S' + \mathbf{q}', && k_S'' = q'', \\ &\mathbf{k}_l' = \mathbf{k}_S^0 + \mathbf{q}^0. \end{aligned} \tag{43}$$

We are interested in the solution $\Delta K \ll k_{Sz}^0, k_{vz}^0$. In this case Eq. (42) reduces to

$$\begin{aligned} (-\Delta K + i\alpha_S)&(\Delta K + F_1/2k_{vz}^0)(\Delta K + F_2/2k_{vz}^0) \\ &+ \Lambda_1(\Delta K + F_2/2k_{vz}^0) - \Lambda_2(\Delta K + F_1/2k_{vz}^0) \\ &+ \Lambda_3(-\Delta K + i\alpha_S) - \Lambda_{123} = 0, \end{aligned} \tag{44}$$

where

$$\begin{aligned} F_1 &= (\omega - \omega_0 - \beta(q^0)^2 - i\Gamma_S)/\beta, \\ F_2 &= (\rho_a/c_a)(\omega^2 - i2\omega\Gamma) - (q^0)^2, \\ \Lambda_1 &= -\lambda_1\lambda_{12}|\mathcal{E}_l|^2/4k_{Sz}^0q_z^0, \\ \Lambda_2 &= \lambda_3\lambda_7|\mathcal{E}_l|^2/4k_{Sz}^0q_z^0, \\ \Lambda_3 &= -\lambda_8\lambda_{11}/4(q_z^0)^2, \\ \Lambda_{123} &= -(\lambda_1\lambda_7\lambda_{11} + \lambda_3\lambda_8\lambda_{12})|\mathcal{E}_l|^2/8k_{Sz}^0(q_z^0)^2. \end{aligned}$$

The solution of Eq. (44) is shown diagrammatically in Fig. 3. Curves 1 and 2 are the dispersion curves for the acoustic wave A_v and the acoustic spin wave $\langle S\rangle$. The momentum-energy matching relation for E_S,

$$q = |\mathbf{k}_l - \mathbf{k}_S| = [\omega_l(n_l\hat{k}_l - n_S\hat{k}_S)\cdot\hat{q}^0 + \omega n_S\hat{k}_S\cdot\hat{q}^0]/c,$$

is given by curve 3. For small coupling constants, the curves simply cross one another instead of forming gaps.[29] Two waves can be coupled effectively only near the point where the corresponding curves meet. Thus, in the parametric approximation stimulated Brillouin

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[29] No gap appears at the junction of phonon and magnon dispersion curves, if $\Lambda_3 \ll (\rho_a/c_a)\omega\Gamma\Gamma_S/\beta k_v^2$. See Ref. 11.

and spin Raman effects will occur essentially independently near the points R_1 and R_2, respectively,[11] where the corresponding stimulated gain would be maximum. The three waves can be effectively coupled together only when the three waves can be made to meet one another at a single point, (R_3). This can be done by selecting the Stokes direction or by tuning the magnon frequency with the applied dc magnetic field. At this point, the gain for the mixed stimulated Brillouin and spin Raman effect would be close to maximum. Thus, the magnon can be excited indirectly by the Brillouin effect or conversely, the acoustic wave can be excited indirectly by the spin Raman effect. This has practical importance in exciting the magnon wave. Individually, the spin Raman effect may have a higher threshold than the Brillouin effect. However, by coupling the magnon waves to the acoustic waves, they can now be excited with a lower threshold via the Brillouin effect. The excited magnon frequency can be tuned by the dc magnetic field, but the direction of the Stokes radiation will also be changed. The exact solution for the case of three waves tightly coupled together should be obtained directly from Eq. (44). Further algebraic details may be found with the methods of Ref. 11.

The coupling of E_S, E_v, and $\langle S\rangle$ can in principle be discussed in the same manner. The nonlinear coupling between E_S and E_v is often small in magnetic media which are nonpiezoelectric. The mixed spin and infrared excitation is more easily excited through the spin Raman coupling. In isotropic media, the dispersion curves for the acoustic magnon and the infrared waves (curves 1 and 2, respectively, in Fig. 4) could not intersect the curve satisfying the linear momentum and energy matching relation at the same point. Therefore, the waves E_S, E_v, and $\langle S\rangle$ cannot be effectively coupled simultaneously. In anisotropic media with indices of refraction satisfying the inequality $n_l < n_S$, this is however, possible as indicated by the resonant point R in Fig. 4. The problem is similar to the Raman effect in polar media discussed by Loudon.[30]

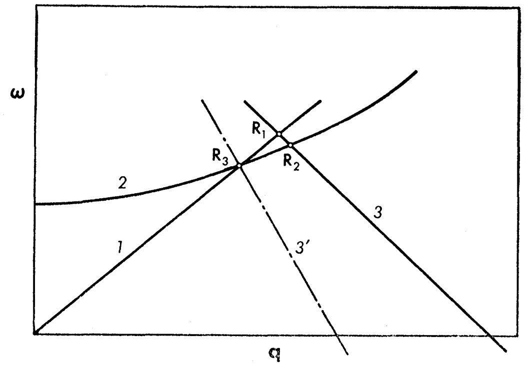

FIG. 3. Dispersion curves describing the mixed spin Raman and Brillouin effect. Curves 1 and 2 are the dispersion curves for the acoustic phonon and the acoustic magnon waves. Curves 3 and 3′ satisfy the linear momentum and energy matching condition

$$q^0 = |\mathbf{k}_l - \mathbf{k}_S{}^0| = [\omega_l (n_l \hat{k}_l - n_S \hat{k}_S{}^0)\cdot\hat{q}^0 + \omega n_S \hat{k}_S{}^0 \cdot \hat{q}^0]/c$$

for two different directions of Stokes scattering. The resonant points R_1 and R_2 denote almost pure Brillouin effect and spin Raman effect, respectively. The resonant point R_3 corresponds to the mixed stimulated Brillouin and spin Raman effect.

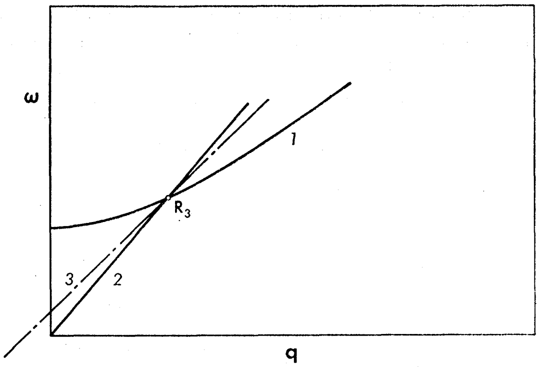

FIG. 4. Dispersion curves describing the simultaneous coupling of the acoustic magnon, the infrared, and the Stokes waves in an anisotropic medium. Curves 1 and 2 are the dispersion curves for the acoustic magnon and the infrared waves, respectively. Curve 3 describes the linear energy and momentum matching condition

$$q^0 = |\mathbf{k}_l - \mathbf{k}_S| = [\omega_l (n_l \hat{k}_l - n_S \hat{k}_S{}^0)\cdot\hat{q}^0 + \omega n_S \hat{k}_S{}^0 \cdot \hat{q}^0]/c$$

in a particular direction of scattering. The resonant point R denotes simultaneous coupling of the three waves.

VI. CONCLUSION

There is a close parallel between the coupling of light with optical phonons and the coupling of light with magnons. The spin Raman process appears to be two orders of magnitude smaller than the ordinary Raman process in liquids. Both the spontaneous and the stimulated spin Raman scattering may be observable in suitable magnetic substances, such as paramagnetic materials at low temperature, insulating antiferromagnets or ferrimagnetic garnets. Magnetic excitations may also be induced by light through the stimulated Brillouin effect and magnetoelastic coupling.

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[30] R. Loudon, Proc. Phys. Soc. (London) **A82**, 393 (1963).