

Effect of Electron Spin Paramagnetism on the Velocity of Sound in Metals

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By means of a simple calculation it is shown that the interaction of an external magnetic field with the magnetic moment of the conduction electrons in a metal gives rise to an increase in the velocity of sound which is independent of the angle between the direction of the magnetic field and the direction of propagation of the sound wave.

THE effect of magnetic fields on the velocity of sound has been studied recently by many authors.¹ The theory predicts an increase in the velocity of sound which depends quadratically on the magnetic field and depends also on the angle between the magnetic field and the direction of propagation of the sound wave. Furthermore, de Haas-van Alphen type oscillations are predicted in the magnetic field dependence. All these effects have been confirmed experimentally.² In this paper we wish to show that the interaction of the magnetic field with the magnetic moment of the conduction electrons also leads to a finite though small increase of the velocity of sound. However, as we will see, this effect is independent of the angle between the direction of the magnetic field and the direction of sound propagation. Since all effects of magnetic fields on the velocity of sound appear through small changes in the dielectric behavior of the conduction electrons, all such changes are additive. Therefore, even for the highest attainable fields, we may concentrate exclusively on the magnetic moment interaction and merely add all previously calculated effects to our final result. We start with the well-known dispersion relation for longitudinal phonons in the random phase approximation (RPA)³

$$\omega^2 = \omega_i^2 / \epsilon(K, \omega), \quad (1)$$

where ω_i is the ionic plasma frequency and ϵ the dielectric constant in the RPA⁴:

$$\epsilon = 1 - \frac{e^2}{\pi^2 m} \int d^3k \frac{F(k)}{[\omega - (h/m)\mathbf{k} \cdot \mathbf{K}]^2 - [(h/2m)K^2]^2}. \quad (2)$$

The Fermi distribution at 0°K is simply given by a step function:

$$F(k) = S(k_F - k), \quad (3)$$

with $k_F = (3\pi^2 N)^{1/3}$ the Fermi momentum divided by \hbar . Upon application of a uniform constant magnetic field

in, say, the z direction the magnetic moment interaction $-\mu\sigma \cdot \mathbf{H}$ (the Pauli term) induces a change in the Fermi distribution. For electrons with spin up, it becomes

$$F_+(k) = S\{[k_F^2 + (2m\mu/h^2)H]^{1/2} - k\} \quad (4)$$

and for electrons with spin down

$$F_-(k) = S\{[k_F^2 - (2m\mu/h^2)H]^{1/2} - k\}. \quad (5)$$

Here μ is the magnetic moment of an electron and H is the magnitude of the applied magnetic field. All we have to do now is to replace $F(k)$ in expression (2) by $\frac{1}{2}F_+(k) + \frac{1}{2}F_-(k)$ in order to obtain the proper dielectric constant in a magnetic field. Of course we did not take into account the influence of the magnetic field on the orbital motion of the electrons. This has been done by Quinn and Rodriguez.¹ But as we mentioned earlier the effects, being small, are simply additive. Substituting then Eqs. (4) and (5) into expression (2) and expanding in powers of H we find to lowest order in H :

$$\epsilon(k, \omega) = 1 + \frac{3\omega_F^2}{v_F^2 K^2} - \frac{2e^2 \mu^2 H^2}{\pi \hbar^3 v_F^3 K^2}. \quad (6)$$

Equation (6) is valid in the long-wavelength limit. We also neglected terms of the order of c_s/v_F , c_s being the velocity of sound and v_F the Fermi velocity, these terms being of the order of 10^{-3} . Inserting Eq. (6) into Eq. (1) we finally obtain:

$$c_s' = c_s [1 + (e^2 \mu^2 / 3\pi \hbar^3 v_F \omega_F^2) H^2], \quad (7)$$

where

$$c_s = (m/3ZM)^{1/2} v_F, \quad (8)$$

with Z the number of conduction electrons per atom. Equation (8) is the well-known Bohm-Staver result for the velocity of sound.³ That the effect is quite small may be seen by actually inserting numbers. It turns out, for instance, that for potassium

$$(c_s' - c_s)/c_s = 4.3 \times 10^{-19} H^2. \quad (9)$$

A change in the velocity of sound of 1 part in 10^7 can be measured at present.⁵ A change of this magnitude

¹ J. J. Quinn and S. Rodriguez, Phys. Rev. Letters **9**, 145 (1962); M. J. Harrison, Phys. Rev. Letters **9**, 299 (1962); M. H. Cohen, M. J. Harrison, and W. A. Harrison, Phys. Rev. **117**, 937 (1960); G. A. Alers and R. T. Swim, Phys. Rev. Letters **11**, 72 (1963).

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⁵ R. L. Forgacs, IRE Trans. Instr. **19**, 359 (1960).

requires a field of 4.8×10^5 G according to (9). Fields of this strength are presently unavailable. Therefore, an increase in accuracy of measurement is required to make the effect observable. For transition metals with their high paramagnetic susceptibilities we expect a much larger effect, however. Owing to their nonspherical

Fermi surfaces, the effect will depend on crystal orientation.

As opposed to the other effects calculated previously,¹ Eq. (7) shows that the effect considered here is independent of the direction of propagation of sound wave.

Structure of Nonlinear Optical Phenomena in Potassium Dihydrogen Phosphate*

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Previously available data on nonlinear optical phenomena in potassium dihydrogen phosphate (KDP) are interpreted together with new data on the linear electrooptic effect. It is shown that second harmonic generation is dominated by energy levels in the ultraviolet ("electronic" levels) whereas the dc and linear electrooptic effects may have contributions due to processes simultaneously dependent on ultraviolet and infrared ("ionic") levels. The contribution of this "electronic-ionic" process to the dc and linear electrooptic effects is less than 50% and may indeed be negligible. The close relationship between the dc and linear electrooptic effects is reexamined and shown to be in better agreement with experiment than previously reported. The limitations and implications of Kleinman's symmetry condition are discussed in the light of recent experimental data.

I. INTRODUCTION

POTASSIUM dihydrogen phosphate (KDP) has been carefully studied in recent experiments with optical second harmonic generation^{1,2} and optical rectification (the dc effect).³ In this work laser sources were required because of the relative minuteness of these nonlinear phenomena. The linear electrooptic effect (or Pockels) effect has also been measured in KDP,⁴ by techniques which utilize conventional light sources. The magnitudes of all of these phenomena in KDP are amongst the largest observed with any crystal.

The purpose of the present paper is to interpret the previously available data on nonlinear optical phenomena in KDP together with new data on the wavelength dependence of the linear electrooptic effect. It is shown that the second harmonic effect is dominated by energy levels in the ultraviolet ("electronic" levels) whereas the dc and the linear electrooptic effects may have contributions due to processes simultaneously dependent on ultraviolet and infrared ("ionic") levels. Uncertainties in the experimental data make it possible to say only that the contribution of this "electronic-ionic" process to the dc and linear electrooptic effects is less than 50% and may indeed be negligible. The remainder

is due to the "electronic-electronic" process which dominates the second harmonic effect.

An important relationship between the dc and linear electrooptic effects was recognized by Armstrong *et al.*⁵ in one of the first detailed theoretical discussions of nonlinear optical phenomena. In the present paper we reexamine this relationship and find that there is even better agreement between theory and experiment than was previously reported.³ Finally, a symmetry condition first proposed by Kleinman⁶ is discussed in terms of the experimental data and the present analysis.

II. SUMMARY OF EXPERIMENTAL DATA

Three phenomena are examined in the present paper.

- (1) The generation of optical second harmonic.
- (2) The dc effect, which is the production of a steady polarization in the crystal by the action of an intense optical electric field.
- (3) The linear electrooptic effect, which is the modification of the refractive indices by a low-frequency (or dc) electric field.

Each of these effects represents an extra polarization produced in the crystal which is described by a third-rank tensor operating on a bilinear function of the electric field amplitudes. The formulation is summarized in Table I.

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