# SECOND-HARMONIC GENERATION BY DAMPED ALFVÉN WAVES AND HELICONS IN ANISOTROPIC SOLID-STATE PLASMAS

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Observation of second-harmonic ($2\omega$) generation and mixing in damped Alfvén-wave propagation in pure bismuth, and in helicon propagation in tellurium-doped bismuth, is reported.

Second-harmonic generation and mixing have been observed for electromagnetic waves propagating parellel to an external magnetic field in anisotropic solid-state plasmas. The above effects were observed for damped Alfvén-wave propagation through pure bismuth[1] and helicon propagation through tellurium-doped bismuth slabs. Coils were used to excite plane waves of the form

$$\vec{b}=\vec{b}_0 e^{i(\omega t-\vec{k}\cdot\vec{r})}, \tag{1}$$

and the nonlinear effects are shown to result from the dependence of the propagation vector on the rf magnetic field of the wave. In addition, large quantum oscillations have been observed in the harmonic and mixed signals as a result of Shubnikov–de Haas oscillations in the magnetoresistance which also depend on the rf magnetic field. This Letter is devoted primarily to a discussion of the damped Alfvén-wave results since the analysis is much less involved for linear polarization than for circular polarization.

It is convenient to express the dispersion relation in terms of the components of the resistivity tensor since the expression for an anisotropic plasma is relatively simple in this formulation.[2] When $\vec{k}\parallel z$,

$$k_\pm^{\,2}=\frac{\mu_0\omega}{2}\left\{\frac{\pm[-4\rho_{xy}\rho_{yx}-(\rho_{xx}-\rho_{yy})^2]^{1/2}-i(\rho_{xx}+\rho_{yy})}{\rho_{xx}\rho_{yy}-\rho_{xy}\rho_{yx}}\right\} \tag{2}$$

in mks units, so that two elliptically polarized modes of propagation are possible. This expression holds when $\rho_{xy}\neq 0$. If $\rho_{xy}=0$, only linearly polarized waves propagate, and

$$k_\pm=k=(1-i)\left(\frac{\mu_0\omega}{\rho_{yy}}\right)^{1/2} \tag{3}$$

for a wave initially polarized in the $x$ direction. Equation (2) applies, for example, to helicon propagation for which $\rho_{xy}\gg\rho_{xx}$ and Eq. (3) can represent Alfvén- or damped Alfvén-wave propagation. Penz has given an expression for the ratio of the field components for conditions corresponding to Eq. (2).[2]

Nonlinear effects may be thought of as arising from the dependence of the $\rho_{ij}$ on the electric and/or magnetic fields of the wave. We neglect here the dependence on electric field and assume that $\rho_{ij}=\rho_{ij}(\vec{B}+\vec{b})$, where $\vec{B}$ is the static magnetic field. If $b\ll B$, the dependence of $\rho_{ij}(\vec{B})$ on $\vec{b}$ is expressed by a Taylor series to first order in $\vec{b}$:

$$\rho_{ij}(\vec{B}+\vec{b})=\rho_{ij}(\vec{B})+S_{ijk}(\vec{B})b_k+A_{ijk}(\vec{B})b_k+\cdots \tag{4}$$

(summation over repeated indices is implied throughout). This expansion, which is linear in $\vec{b}$, will, when substituted in the dispersion relations, yield the second harmonic in the small-signal case. It is convenient to separate the third-rank tensor coefficient into a symmetric part, $S_{ijk}(\vec{B})=S_{jik}(\vec{B})$, and an antisymmetric part, $A_{ijk}(\vec{B})=-A_{jik}(\vec{B})$, because they transform differently under the symmetry operations.

In the limit $\vec{B}=0$, we have $S_{ijk}(0)=0$ and the $A_{ijk}$ become the weak-field Hall coefficients. When $\vec{B}$ is along a general direction in the crystal, the symmetric and antisymmetric tensors have 18 and nine nonvanishing coefficients, respectively, but application of the symmetry operations for $\vec{B}$ along the principal axes reduces this number considerably. If we further require $\vec{B},\vec{k}\parallel z$, then our attention can be restricted to coefficients of the type $A_{ijj}(B)$ and $S_{ijj}(B)$ $(i,j\neq z)$. For $z$ along the binary (1), bisectrix (2), and trigonal (3) directions of bismuth, there are zero, four, and one such coefficients, respectively.[3] For $\vec{B},\vec{k}\parallel 3$ the applicable expansions are

$$\rho_{11}=\rho_{11}{}^{0}-S_{222}b_2,\quad \rho_{12}=\rho_{12}{}^{0}-S_{222}b_1,$$
$$\rho_{22}=\rho_{11}{}^{0}+S_{222}b_2,\quad \rho_{21}=\rho_{12}{}^{0}-S_{222}b_1. \tag{5}$$

The results for $\vec{B},\vec{k}\parallel 2$ are more complicated and will not be given, but the pertinent nonvanishing coefficients are listed in Table I.

The resistivity tensor and the third-rank tensor coefficients can be evaluated using standard assumptions from the usual Fermi-surface model, and the results for $\rho_{ij}\simeq -i(\epsilon^{-1})_{ij}/\omega\epsilon_0$ are well known.[4] The expressions for the nonlinear coefficients have been derived only for $\vec{B},\vec{k}\parallel 3$. We assume, using standard notation, $\tau_e=\tau_h=\tau$, $\tau$ isotropic, $\omega_c\tau\ll 1, \hbar\omega_c\ll k_BT, v_F\tau k\ll 1, m_1\ll m_2$. The result is

$$S_{222}=-\frac{\rho_{11}{}^{0}}{B}\frac{m_4}{m_3}\left[1+2\frac{p}{n}\left(\frac{M_1}{m_2-m_4{}^2/m_3}\right)\right]^{-1}. \tag{6}$$

Note that this coefficient is proportional to the off-diagonal component of the electron-mass tensor, $m_4\equiv m_{23}$, which is nonzero because the electron Fermi ellipsoids are tilted out of the trigonal plane. Since the corresponding component for holes, $M_4$, is zero, the nonlinear coefficient vanishes when the electron concentration $n$ goes to zero.

Typical experimental results for harmonic generation by a damped Alfvén wave are shown in Fig. 1 for $\vec{b}\parallel 2$. A very similar curve was obtained for $\vec{b}\parallel 1$; however, the harmonic was again polarized $\parallel 2$. Large quantum oscillations due to holes were observed for both orientations, although the oscillations are barely visible in the damped Alfvén transmission. The results for these orientations were interpreted by substituting Eqs. (5) in Eq. (3) when $\vec{b}\parallel 2$ and in Eq. (2) when $\vec{b}\parallel 1$, since $\rho_{xy}\neq 0$ in this case. In both cases this substitution leads to a linear dependence of $k$ on $b$ (higher orders in $b$ are neglected). When this linear term is small, the exponential in Eq. (1) can be expanded to give the harmonic contribution when $b\parallel 2$, as

$$b_H=-ib_0{}^2(S_{222}k_0z/2\rho_{11}{}^0)e^{i(2\omega t-2k_0z)}, \tag{7}$$

where $k_0\equiv(1-i)(\mu_0\omega/\rho_{11}{}^0)^{1/2}$. The result for $\vec{b}\parallel 1$ is the negative of Eq. (7). The polarization of the harmonic when $\vec{b}\parallel 2$ is the same as the fundamental since Eq. (3) applies. When $\vec{b}\parallel 1$, the $k_+$ and $k_-$ solutions of Eq. (2) must be combined to satisfy the initial boundary conditions. If this is done (the expression given by Penz for the ratio of the field components is used), the harmonic is found to be polarized at right angles to the fundamental in agreement with experiment.

Equation (7) predicts a maximum in the harmonic signal as a function of $B$ as was observed experimentally. In the magnetic field region of interest, $\rho_{11}{}^0\propto B^2$, and therefore, using Eqs. (6) and (7), $|b_H|\propto B^{-2}\exp(-\text{const}/B)$. Points calculated using Eq. (7) are represented in Fig. 1 by triangles. The solution arrived at in Eq. (7) does not take into account the fact that once the harmonic is generated at $2\omega$, it propagates as a damped Alfvén wave with a wave vector $(2k_0)^{1/2}$. Interference effects in the generated signal resulting from this phase mismatch were taken into account in calculating the solid circles, and the agreement with experiment is somewhat better. Internal reflections were neglected in both cases. The theoretical magnitude of the effect can be estimated from Eq. (7) if $b_0$ is known.

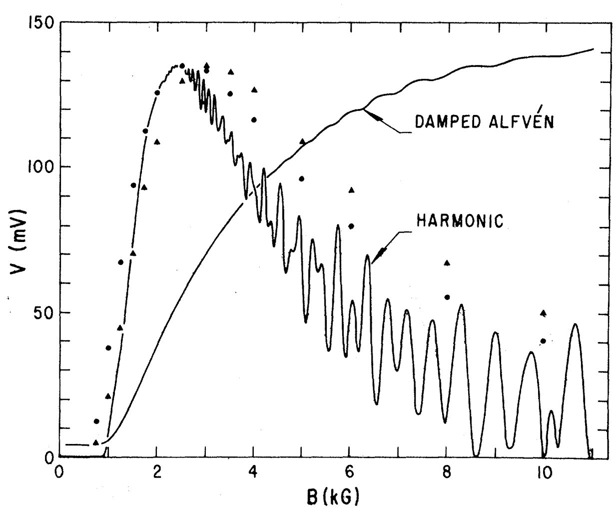


FIG. 1. Amplitude (times $3\times10^4$) of 30-MHz harmonic generated by 15-MHz damped Alfvén wave (also shown) as a function of magnetic field at 1.2°K. The magnetic field and propagation directions were $\parallel 3$, and both signals were polarized $\parallel 2$. The triangles were calculated from Eq. (7) using the effective masses determined by Y. H. Kao, Phys. Rev. 129, 1122 (1963), and $\tau_e=\tau_h=2.65\times10^{-10}$ sec which fits the damped Alfvén transmission. The solid circles were calculated including a correction for phase mismatch. In both cases the points were normalized at the peak of the harmonic curve.

The value of $b_0$ is estimated to be ~3 G from the damped Alfvén data. This leads to an estimated value of the effect at the peak approximately 10 times greater than observed experimentally. It was experimentally determined that the amplitude of the harmonic signal was proportional to the square of the amplitude of the fundamental in agreement with Eq. (7).

Damped Alfvén-wave results for all principal orientations are given in Table I with $\vec{k} \parallel \vec{B}$ in all cases. The comparisons were made at the magnetic field corresponding to the peak in the harmonic for the given field orientation, and $b_0$ was the same for all cases. For some rf field orientations a distinct peak was not observed, although quantum oscillations were always observed. The nonvanishing expansion coefficients which can lead to an effect are listed in Table I. There were two orientations for which the observed effect was relatively large when no effect should have been observed according to the preceding symmetry arguments. This is probably a result of misorientation of the sample and the rf coils (see comment in Table I). The case $\vec{B}, k \parallel 2$ is quite complicated with regard both to evaluation of the coefficient and to application of the results to the model. The experimental results indicate that $S_{311}$ is the dominant coefficient.

The large quantum oscillations which have been observed for all orientations are generally small in the region of the peak in the harmonic. The oscillations shown in Fig. 1 grow in magnitude with magnetic field. For some orientations (e.g., $\vec{B}, \vec{k} \parallel 2$; $\vec{b}, \vec{b}_H \parallel 1$) the oscillations reach a maximum at 6 kG and decrease in amplitude at higher fields. This is not surprising because $|b_H| \propto B^{-2} \exp(-\mathrm{const}/B)$ and the amplitude of the Shubnikov–de Haas oscillations saturates at large $B$. Therefore the amplitude of the oscillation in the harmonic should decrease for large $B$. Although the second harmonic signal results from the tilt of the electron ellipsoids, the appearance of small hole oscillations would be expected because of the presence of $k_0$ in Eq. (7). An explanation of the giant oscillations actually observed would require a quantum-mechanical calculation.

The periods of all observed oscillations are in good agreement with recent de Haas–Van Alphen data.[6] Oscillations with half the de Haas–Van Al-

Table I. Summary of damped Alfvén-wave results.

| Sample [a] | Magnetic Field Directions | | | $2\omega$ Harmonic ($\mu$V) | Pertinent Nonvanishing Coefficients | Comments |
|---|---|---|---|---|---|---|
| | $\underline{B}$ | $\underline{b}$ | $\underline{b}_H$ | | | |
| Bi 77-2 | 1 | 2 | 2 | 2.16 | None | $30^\circ$ from $\underline{B} \parallel 2$; $\underline{b}$, $\underline{b}_H \parallel 1$. |
| 2.14 mm | | | 3 | 0.60 | None | |
| 2.16 kG | | 3 | 2 | 0.20 | None | |
| | | | 3 | 0.32 | None | |
| Bi 77-4 | 2 | 1 | 1 | 4.70 | $S_{311}$ | The case $\underline{B}$ $\underline{k} \parallel 2$ is complicated |
| 2.03 mm | | | 3 | 0.95 | $S_{311}$ | because $\rho_{13}^0 \neq 0$ and $\rho_{11}^0 \neq \rho_{33}^0$ . |
| 1.58 kG | | 3 | 1 | 0.90 | $S_{113}$, $S_{333}$, $A_{313}$ | |
| | | | 3 | 1.07 | $S_{113}$, $S_{333}$, $A_{313}$ | |
| Bi 75-4 | 3 | 1 | 1 | 1.67 | None | $30^\circ$ from $\underline{B} \parallel 3$; $\underline{b}$, $\underline{b}_H \parallel 2$ . |
| 1.97 mm | | | 2 | 4.02 | $S_{222}$ | |
| 2.25 kG | | 2 | 1 | 0.03 | None | |
| | | | 2 | 4.45 | $S_{222}$ | |

[a]Slab thickness and magnetic fields corresponding to listed harmonic voltages are also given.

phen period were also observed in most cases, although they were generally not present in the transmission curve for the fundamental.

All of the above-mentioned results are supported by results obtained by mixing two damped Alfvén waves at 12.5 and 17.5 MHz to produce a signal at 30.0 MHz. The mixing data are complicated by an experimental problem of achieving $\vec{k} \parallel \vec{B}$ for both signals and by a more severe problem with phase mismatch, so that the agreement with theory is not quite as good.

Harmonic generation and mixing have also been observed for helicon propagation through tellurium-doped bismuth slabs with $n \approx 2 \times 10^{18}$ $cm^{-3}$ for $\vec{k}$ and $\vec{B}$ in the three principal directions. Analysis of these results is complicated by phase mismatch and the fact that reflections cannot be ignored since Fabry-Perot peaks are observed in the helicon transmission curve. Equation (2) with $\rho_{xy} \gg \rho_{xx}$ is applicable for this case, but because of the above complications the results have not yet been analyzed. An apparent Fabry-Perot peak in the harmonic was observed for $\vec{k}, \vec{B} \parallel 3$, and the signal was increasing at the highest magnetic field achieved (11 kG). The effects for $\vec{k}, \vec{B} \parallel 1$ and 2 seemed to be completely due to quantum oscillations.

The phenomenological description employed here assumes that the nonlinear effects arise in this particular case primarily from the interaction of the magnetic field of the wave with the medium. Since the observed dependence of the harmonic signal on the polarization of the wave is as predicted, the assumption appears to be valid. If the electric field were responsible for the effect, the polarization dependence would be just the opposite of that actually observed. The appearance of signals in some of the cases where none is predicted could, of course, be the result of an electric-field-induced nonlinearity. However, since the dc current-voltage characteristics reveal only magnetic-field-induced effects,[7] it is our opinion that violation of the conditions $\vec{B}, \vec{k} \parallel$ (principal axis) is a more plausible explanation.

The agreement of the order of magnitude and the magnetic field dependence of the effect with the approximate theory derived herein suggests that the mechanism of generation has also been correctly identified. It follows that the effect should be expected in semimentals and semiconductors having anisotropic many-valley band structures. Provided the stated assumptions are valid, Eqs. (6) and (7) hold for arbitrary $\omega\tau$ and therefore also apply to undamped Alfvén waves. The symmetry arguments are, of course, even more general.

For the case of cubic crystals possessing inversion symmetry, the nonvanishing tensor coefficients in the presence of an external magnetic field are the same as for bismuth (point group $\bar{3}m$), provided axes 1, 2, and 3 are taken as $[\bar{1}01]$, $[1\bar{2}1]$, and $[111]$ axes, respectively. Thus, although no harmonic generation at $2\omega$ is allowed for $\vec{B}$ and $\vec{k}$ along the cubic axes, an effect described by Eqs. (5) is expected for $\vec{B}$ and $\vec{k}$ along a $[111]$ axis.

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