Nuclear Electric-Quadrupole Radiative Transitions as an Explanation of InSb Microwave Emission

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InSb samples at 4 K and $B_{ext}=0$ are observed to emit narrow-band radiation in two bands near 400 and 600 MHz. The frequency ratio of the two bands strongly suggests that the emission is due to nuclear electric-quadrupole transitions $\frac{3}{2}\to\frac{1}{2}$ of Sb^{121} and Sb^{123} nuclei located near impurity atoms. The nuclear electric quadrupole hypothesis explains simply many of the previously puzzling aspects of Insb microwave emission.

This paper presents evidence to show that some of the radio and microwave frequency emission from InSb[1-9] is due to nuclear electric-quadrupole (NEQ) transitions of the In and Sb nuclei, and that this hypothesis explains many of the properties of the emission which were not understood previously.

Larrabee and Hicinbothen[1,2] first observed microwave emission from InSb at 77 K when it was subjected to strong electric and magnetic fields. Although Buchsbaum, Chynoweth, and Feldmann[3] reported that emission could be observed in weak electric fields, Thompson and Kino[4] demonstrated that most, if not all, of the "low-field emission" is from small regions with large electric fields created near the contacts. They further indicated that avalanche of the carriers is necessary for emission. In addition, some zero-magnetic-field emission is reported.[5] The frequency spectrum of emission has been observed to consist of a diversity of types from narrow-band, coherent to broad-band noise extending from 10 MHz to $\gtrsim 100$ GHz. Many explanations have been advanced to explain the observations, including helicons, phonon generation, plasma instabilities, and amplification of shot noise. However, many aspects of the InSb emission are not understood; in particular, the observation of several series of lines in the region between 20 and 7000 MHz reported by a number of authors.[6-9] These lines may be typically characterized as a series of four lines whose "fundamental" frequency is in the vicinity of 50 MHz (though in fact, observed from 22 to 90 MHz), and another line usually observed between 400 and 600 MHz, sometimes possessing a "harmonic."

The InSb (and the BiSb[10]) emission lines and properties suggest energy levels, but no obvious ones presented themselves until it was established[11] that line-narrowed radiation was indeed being emitted from BiSb, and further it was probable that the energy levels existed even with zero magnetic field. NEQ levels have this property and suggest several tests of this hypothesis. Some of these tests are reported here for the InSb emission.

An atomic nucleus possesses NEQ energy levels if (1) the nucleus has a nuclear electric-quadrupole moment Q and (2) occupies a site with an electric field gradient $\nabla\vec{E}$. The Hamiltonian of such a system is given by $H=Q_{ij}\nabla_i E_j$ where $\overleftrightarrow{Q}$ and $\nabla\vec{E}$ are symmetric second-rank tensors. The NEQ energy levels $E(m)$, degenerate in $\pm m$, the axial component of the nuclear spin I, are given by

$$E(m)=\frac{e^2qQ}{4I(2I-1)}[3m^2-I(I+1)],$$

where q is proportional to the electric field gradient. Transitions are permitted which satisfy the selection rule $\Delta m=\pm1$. [Nuclear Zeeman, hyperfine interactions, and departures from axially symmetric electric field gradients modify Eq. (1), but for simplicity these are ignored here.]

InSb has a cubic (zinc-blende) crystal structure, which forbids an electric field gradient at the nucleus of a perfect crystal. However, if impurities (or other imperfections) are introduced, the nuclei in their vicinity can experience electric field gradients which lead to NEQ energy levels. In InSb, impurities are known to be present at concentrations of $\sim10^{15}/cm^3$ even in the purest samples. We assume here that these impurities are the source of the electric field gradients required for NEQ levels in InSb. Indeed, it will be seen that this assumption leads to a very simple explanation of some of the most peculiar properties of the InSb radio-frequency emission. Since no NEQ resonances of In or Sb due to impurities have been measured in InSb, we must use other means than a direct comparison of frequencies to test the NEQ hypothesis.

The relevant nuclear properties of the In and

TABLE I. Nuclear properties of In and Sb isotopes.

Nuclear species	Natural abundance	I	Q (10^{-24} cm^2)	$\frac{\lvert Q\rvert}{4I(2I-1)}$ [a]
In^{113}	4	9/2	0.75	0.644
In^{115}	96	9/2	0.761	0.653
Sb^{121}	57	5/2	−0.53	1.647
Sb^{123}	43	7/2	−0.68	1.000

[a]Normalized to Sb^{123} value.

Sb isotopes are listed in Table I. This table gives the isotopic species, its natural abundance, nuclear spin I, and quadrupole moment Q. The last column gives the value $Q/4I(2I-1)$ normalized to the Sb^{123} nucleus, which is proportional to the frequency of a specific NEQ transition in a given electric field gradient.

The InSb system, according to Eq. (1), has five NEQ levels and four allowed NEQ transitions for the In^{115} (and also In^{113}) nucleus; i.e., $\frac{1}{2}\leftrightarrow\frac{3}{2}$, $\frac{3}{2}\leftrightarrow\frac{5}{2}$, $\frac{5}{2}\leftrightarrow\frac{7}{2}$, and $\frac{7}{2}\leftrightarrow\frac{9}{2}$. Furthermore, these are approximately in the frequency ratio of 1:2:3:4. Similarly, the dominant (57% abundance) antimony isotope Sb^{121} with $I=\frac{5}{2}$ shows two NEQ transitions with a frequency ratio of 1:2 and the Sb^{123} with $I=\frac{7}{2}$ shows three transitions with ratios 1:2:3.

Examining InSb emission data[6-9] in the light of possible radiative NEQ transitions, it immediately becomes suggestive to identify the four low-frequency lines (in the region of 20 to 300 MHz) and the one or two high-frequency lines (400 to 1000 MHz) with the In^{115} ($I=\frac{9}{2}$) and the Sb^{121} ($I=\frac{5}{2}$) NEQ transitions, respectively. However, it is most unlikely that the In^{115} and Sb^{121} nuclei see identical electric field gradients, so that for a similar transition (say $\frac{3}{2}\leftrightarrow\frac{1}{2}$), the frequencies are not expected to be in the ratio of 2.52, as would be predicted by Table I, were that the case.

Nevertheless, each antimony nuclear species, Sb^{121} and Sb^{123}, should see identical electric field gradients for a specific impurity. This suggests that we look below the postulated Sb^{121} emission line for the emission of a frequency at 1/1.647 (see Table I) times the Sb^{121} frequency which would be associated with the Sb^{123} nuclear species. Indeed, such a line is found and the pair is discussed below.

[The postulated NEQ frequencies are rather high, but they lie within the range known to exist in other materials. Furthermore, the Sternheimer antishielding factor in InSb[12] is unusually large (~1000), and this enhances impurity-induced electric field gradients and therefore NEQ frequencies by a similar amount.]

Current pulses (3 μsec long and at a 100-Hz rate) were applied to an n-type InSb sample ($n \approx 10^{14}/\text{cm}^3$), mounted near the termination of a coaxial line, and immersed in liquid helium. The dc voltage of the current pulse was blocked from the detection system by a high-pass filter. In addition, a low-pass and a narrow-band, band-pass filter were used to avoid spurious signals. The preamplified signal was detected by a superhetrodyne receiver.

When a strong magnetic field was applied, the emission had threshold curves similar to those previously reported[1-9] for InSb. In addition, four lines were observed near 57, 124, 186, and 244 MHz, similar to those previously reported.[6-9] Several high-frequency lines were observed in the 300 to 24 000-MHz region. The frequencies of the strongest pair of lines (near 490 and 820 MHz with $B=5$ kG, for example) were found to have a frequency ratio of 1.67 ± 0.03, supporting the hypothesis that these lines are due to the $\frac{3}{2}\leftrightarrow\frac{1}{2}$ NEQ transitions of the Sb^{123} and Sb^{121} nuclei, respectively.

In addition, it was discovered that the InSb samples at 4 K emitted narrow-band radiation in bands in *zero* external magnetic field, similar to the BiSb emission,[10,11] and also tuning with current in a similar manner. A pair of these emission bands (curves 1 and 2) is shown in Fig. 1, where their frequency is given as a function of sample current. Some emission is also observed in other parts of the f-versus-I plane, as well as a "harmonic" of the 600-MHz band; however, only the two "fundamental" bands are shown.

The bands (curves 1 and 2) in Fig. 1 (labeled by the Sb^{121} and Sb^{123} isotopes and the NEQ transitions thought to be responsible) have been observed with $B_{\text{ext}}=0$ in each of the three InSb samples investigated to date. Although the dimensions and electrical conductivity of the samples vary by nearly an order of magnitude, and both rectifying and nonrectifying contacts have been employed, each of the three samples have shown two bands with frequencies close to those of Fig. 1, and the bands of all samples tune similarly. Curve 3 shows the ratio of the frequencies of the two bands, which is expected to be about 1.65 if the lines are due to the assigned transitions. This curve shows deviations from about +6 to −15% from the theoretical value, with an average value of 1.61. Despite the small departure from the theoretical value, it is believed that

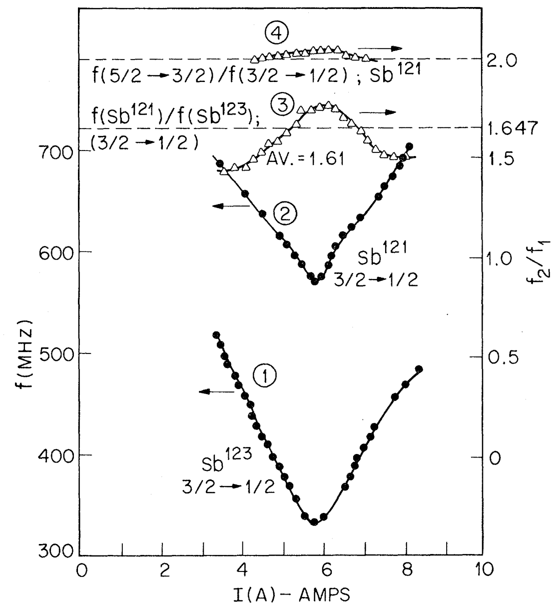

FIG. 1. Two emission bands observed from an n-type InSb sample at 4 K as a function of current and $B_{ext}=0$ (curves 1 and 2). The sample has indium contacts and dimensions of $1.19\times 1.30\times 11.4$ mm^3. The labels denote the attributed NEQ radiative transitions. Curves 3 and 4 show the ratio of the frequencies of various bands of the InSb $(N-1)$ emission as a function of current. The horizontal dashed lines correspond to theoretical frequency ratios of the NEQ transitions labeled.

these are indeed the labeled transitions, because the very substantial tuning (~60%) apparently tunes each species of nuclear spin somewhat differently.

The tuning mechanism (discussed in detail elsewhere[12]) of the zero–magnetic-field emission is undoubtedly complex. The tuning definitely involves the self–magnetic-field and probably hyperfine effects, and a radial shift of the emitting regions inside the sample. The present understanding of the tuning predicts qualitatively the behavior of the ratio as observed in Fig. 1 for the case of zero external magnetic field. In the case of large external magnetic field ($\gg B_{self}$), all nuclei see the same magnetic field, and the ratio is expected to be close to the theoretical value. This is in fact found to be the case.

It becomes less surprising that different nuclear species tune differently, when the observation is made that the frequency ratio of different levels of the *same* species departs from their theoretical integer ratio in a manner similar to the different species, although more weakly. This is illustrated in curve 4 of Fig. 1 where the frequency ratio of the transitions $(\frac{5}{2}\to\frac{3}{2})/(\frac{3}{2}\to\frac{1}{2})$ is shown for the Sb^{121} nuclei. [The Sb^{123} $(\frac{3}{2}\to\frac{1}{2})$ transition is already weak, so that no higher-level transitions are observed here.]

The frequency ratio of these two bands (Sb^{121} and Sb^{123}) in the other InSb samples has a shape similar to that shown in curve 3. The average ratio of these two bands for all three samples is 1.62, or less than 2% from the theoretical value of 1.647.

Arizumi *et al.*[7] report InSb emission in bands between 60 and 280 MHz, as well as two bands near 4 and 7 GHz, whose maxima are at approximately 4.3 and 7.2 GHz. The ratio of these frequencies is 1.67 or a value close to the Sb^{121} and Sb^{123} quadrupole frequency ratio. Assuming these are NEQ levels, the higher-frequency transitions lie at 8.6, 12.9, and 14.4 GHz. Unfortunately, Arizumi's data do not extend beyond ~7.5 GHz, so a test is not possible using his data. However, Suzuki's[13] InSb emission data range from 7.0 to 26 GHz. His first four emission peaks (also the strongest ones) lie at about 7.2 (Arimuzi's[7] higher frequency), 8.8, 13.2, and 15 GHz, i.e., within a few percent of the NEQ predicted values. It is not unreasonable that Arizumi's and Suzuki's different samples, both of high purity, are exhibiting NEQ levels due to the same impurity, since certain impurities are difficult to remove and appear in all InSb. Frequencies close to these have also been observed (unpublished) by the author from high-purity InSb.

Most of the high–magnetic-field InSb emission lines[6-9] tune with electric and magnetic fields as does the zero field emission. We attribute this tuning to the effect of ionized (i.e., *charged*) impurities responsible for the quadrupolar splitting. The magnetic and electric fields affect both the localization of, and hyperfine splitting at, the nuclear levels due to the residual charge. Both effects alter the NEQ levels.

Several workers[7,8] report four InSb emission lines whose "fundamental" is near 80 MHz and whose frequencies are *independent* of magnetic field. We speculate that these lines are due to quadrupolar splitting introduced by a neutral impurity (e.g., As) at an Sb lattice site. In this case there is no residual charge for the magnetic field to influence. The above speculations are in agreement with the well-known[14] fact that charged impurities in InSb create an order-of-magnitude greater quadrupolar broadening of NMR levels than do neutral impurities.

As one further example of simplification of previously complex InSb data, consider the work of Tacano and Kataoka (last paper of Ref. 8), who report two "modes" of emission with fundamentals near 400 MHz (two lines) and 20 MHz (four lines), and whose frequencies increase and decrease with B, respectively. A simple understanding results if we assume that the impurity is a charged one whose charge is localized with B (a well-known effect), and the modes are due to NEQ transitions of first nearest neighbors (Sb^{121}) and second nearest neighbors (In^{115}). This hypothesis predicts the number of lines observed and the frequency ratio of the lines and the two "modes." The hypothesis furthermore leads to a simple explanation of the modes' opposite tuning in B.

In conclusion, the evidence strongly suggests that most of the lines of emission from InSb are due to NEQ radiative transitions of the In^{115}, Sb^{121}, and Sb^{123} nuclei located in the vicinity of impurity atoms. The zero-field, narrow-band emission observed at 4 K from InSb is believed to be stimulated emission and pumped with the same avalanche mechanism as the BiSb stimulated emission.[11] The large-B NEQ emission lines at 77 K are presumably pumped in a similar manner, and some aspects of the InSb pumping are discussed in Ref. 11. Present indications are that the InSb *noise* emission is also in part due to spin transitions, perhaps a combination of electron, nuclear magnetic, and nuclear quadrupole transitions. The identification of strong NEQ emission from lightly doped InSb suggests many novel and previously inaccessible studies of this and other materials.

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Continuous Line-Narrowed, Coherent Radiation from Pumped $Bi_{92}Sb_{08}$ Nuclear Electric-Quadrupole Levels

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We report the observation of strong line narrowing of microwave radiation (870 MHz) emitted from a continuously operated, dc-pumped $Bi_{92}Sb_{08}$ semiconducting alloy at 1.6 K. Bandwidths of less than 1 kHz are observed. The origin of the line narrowing is thought to be a maser effect involving the $\frac{3}{2}\rightarrow\frac{1}{2}$ nuclear electric-quadrupole transition of the Bi^{209} nucleus. A new dc pumping mechanism is proposed to explain the effect. In this effect the nuclei are pumped by electronic spin levels, saturated as in the Overhauser effect, except that saturation is achieved by carrier avalanche, instead of rf pumping.

The emission of extremely narrow-band, coherent microwave radiation[1] from BiSb alloys in zero external magnetic field has not been explained. This radiation is observed in four discrete, nearly harmonic bands, with the "fundamental" near 1000 MHz, and is coherent in all four bands. The radiation is excited by a pulsed dc current. It has been further shown[2] that broadband noise is also emitted from BiSb, and that the onset of both types of emission is associated