COMMENTS AND ADDENDA

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Use of the Two-Frequency Technique of Prior *et al.* for hfs Investigations of Atoms with Integral J*

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It is shown that in using the two-frequency technique of Prior *et al.* for investigation of the hyperfine structure of atoms with integral J, any resonance observed may arise from a two-quantum transition ($\nu_\alpha + \nu_\beta = \Delta E/h$; $\nu_\alpha \neq \nu_\beta$) rather than from two successive single-quantum jumps. Although this feature requires that care be used in interpreting the observed resonance frequencies, it appears to extend the usefulness of the technique as a spectroscopic tool.

I. INTRODUCTION

The procedure for conducting atomic-beam magnetic-resonance investigations of hyperfine structure (hfs) in atoms with half-integral J is well established. One or more $\Delta F = 0$, $\Delta M_F = \pm 1$ transitions between states for which $M_J = +\frac{1}{2}$ and $-\frac{1}{2}$ at high field are followed in frequency to higher and higher magnetic fields until the resonance frequencies for the $\Delta_F = \pm 1$, $\Delta M_F = 0, \pm 1$ transitions can be predicted with sufficient accuracy at low field. Subsequent observation of these "direct" transitions then completes the measurements. The transitions observed are all single-quantum magnetic-dipole ones, and the transition probability is normally appreciable at modest rf power levels. The large number of such experiments which have been successfully performed on radioactive atoms is evidence of the good signal-to-noise ratio commonly attainable for low-lying atomic states.

The situation is basically different for atoms with integral J, however. This is illustrated in Fig. 1(a), a schematic hyperfine-structure diagram for an atom with $J = 1$ and a nuclear spin $I = \frac{1}{2}$. The three levels of particular interest are shown (for a moderate value of the field) in an expanded scale in Fig. 1(b) and the appropriate quantum numbers are indicated. As can be seen, there can be no $\Delta_F = 0$, $\Delta M_F = \pm 1$ transitions between levels that have equal and opposite values of M_J at high field. One approach to the problem has been to remove the usual central obstacle in the atomic-beam magnetic-resonance apparatus and to work with $\Delta F = 0$, $\Delta M_F = \pm 1$ "flop-out" transitions (such as ν_1 and ν_2 in Fig. 1) between states having $M_J = 0$ and $M_J = +1$ or -1 at high field. While these transitions are single quantum and easy to induce, they can be difficult to observe because they produce only a very small decrease in a large background beam (consisting mostly of atoms with $M_J = 0$). The very small signal-to-background ratio may not preclude such observations on beams of stable atoms if a universal detector is used, but it usually does make it impossible to use these transitions in work with radioactive beams using deposition detection techniques.

The technique most commonly used for work on atoms of integral J, particularly for radioactive beams for which the flop-out transitions are so difficult to observe, is to apply sufficient rf power to induce $\Delta_F = 0$, $\Delta M_F = 2, 4, 6, \ldots$ multiple-quantum transitions[1] between states having equal and opposite values of M_J at high field [such as $F = \frac{3}{2}$, $M_F = \frac{1}{2} \rightarrow F = \frac{3}{2}$, $M_F = -\frac{3}{2}$, illustrated in the center of Fig. 1(b)]. This technique permits use of a central obstacle to block undeflected atoms and can lead to good signal-to-noise ratios. The principal disadvantage of the technique is that as the magnetic field is increased, the $\Delta M_F = \pm 1$ energy spacings become more and more unequal, with a rapid resul-

tant drop in transition probability. Although one can make corrections[2] for the "frequency pulling" which may be caused by the high rf power required to induce the transitions, the situation becomes increasingly troublesome as the field is increased.

A different technique for investigation of the hfs of an atom with integral J has been used by Prior *et al.*[3] to study the $^3P_1$ metastable atomic state in several radioactive Sn isotopes. It involves the simultaneous application of two rf signals ν_α and ν_β to enable the atoms of the beam to make two successive single-quantum jumps. The two frequencies are chosen such that ν_α is the energy difference at field H between states having $M_J = +1$ and 0 at high field, and ν_β is the energy difference between states having $M_J = 0$ and -1 at high field; i.e., if the method were to be used on the atom of Fig. 1, one would choose $\nu_\alpha = \nu_1$ and $\nu_\beta = \nu_2$. With this arrangement, a central obstacle can be used, and enough atoms can make the two-quantum jump (by interacting with the two quanta successively) to produce a flop-in signal with a good signal-to-noise ratio. An obvious disadvantage of the scheme is that each of the two rf frequencies must be very near the appropriate single-quantum energy interval if the transition probability is to be appreciably different from zero. The great advantage is that the technique is applicable at arbitrary field and, since it requires only modest rf power, is less likely to lead to severe pulling.

The purpose of this comment is to point out an additional feature of the technique, a feature which can be either an advantage or a disadvantage. It has been known for some time[2,4] that there can be appreciable transition probability for an atomic multiple-quantum transition even if several of the quanta have frequencies which are very different. For the atom of Fig. 1 in particular, if we simultaneously apply the two frequencies $\nu_\alpha = \nu_1 - \epsilon$ and $\nu_\beta = \nu_2 + \epsilon$, we can expect many atoms to undergo the transition $(\frac{3}{2}, \frac{1}{2} \leftrightarrow \frac{3}{2}, -\frac{3}{2})$ for a wide range of values of ϵ. Thus, a resonance observed when two frequencies are applied is not necessarily due to two successive single-quantum jumps as anticipated, and the conclusions concerning the hfs constants and g_J value of the state being studied must be drawn accordingly. The implications of this ambiguity will be discussed in Sec. III, but first these points will be illustrated by a very brief report on the results of some measurements performed on the $^3P_1$ metastable state of Sn^{119}.

II. MEASUREMENTS

The Zeeman's effect and the hyperfine structure of the $^3P_1$ metastable atomic state of Sn^{119} were studied in detail by Childs and Goodman,[5] and the transition frequency for any transition can be accurately calculated for a given value of the field by standard techniques. Figure 1(a) indicates the hyperfine structure schematically ($I = \frac{1}{2}$ for Sn^{119}) as a function of the field, and Fig. 1(b) shows the situation for the levels of interest at $H = 100$ G, at which the present observations were made. At 100 G, it is known that

$$\nu_1 \equiv [E(\tfrac{3}{2}, \tfrac{1}{2}) - E(\tfrac{3}{2}, -\tfrac{1}{2})]/h = 141.888(7) \text{ MHz},$$

$$\nu_2 \equiv [E(\tfrac{3}{2}, -\tfrac{1}{2}) - E(\tfrac{3}{2}, -\tfrac{3}{2})]/h = 150.458(8) \text{ MHz},$$

from which it follows that the normal double-quantum frequency (the term "normal double-quantum transition" will be used to denote one for which the two quanta are identical) is

$$\nu_{2Q} = \tfrac{1}{2}[E(\tfrac{3}{2}, \tfrac{1}{2}) - E(\tfrac{3}{2}, -\tfrac{3}{2})]/h \doteq \tfrac{1}{2}(\nu_1 + \nu_2)$$
$$= 146.173(6) \text{ MHz}.$$

An Anzac Electronics model H-8 hybrid junction was arranged to permit simultaneous application

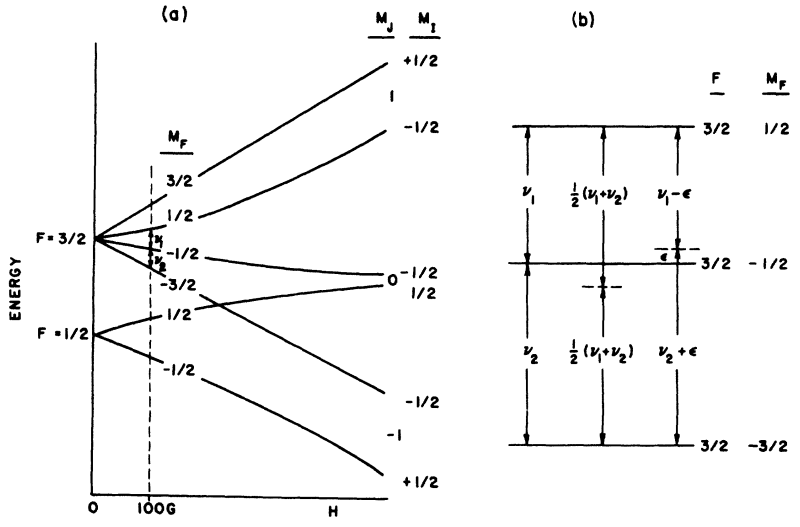

FIG. 1. Schematic hyperfine-structure diagrams showing transitions of the type discussed. (a) Magnetic field dependence of the hyperfine structure of the $^3P_1$ metastable atomic state of Sn^{119}. (b) Three types of transitions involving the $F = \frac{3}{2}$, $M_F = \frac{1}{2}, -\frac{1}{2}, -\frac{3}{2}$ levels at 100 G, the magnetic field at which the experiment was performed. The two quanta required for the change of state $M_F = \frac{1}{2} \leftrightarrow -\frac{3}{2}$ can be chosen in any of the three ways indicated. The mechanisms at the left and center require well-specified discrete frequencies. For the two-quantum scheme at the right, however, the quantity ϵ can be as large as 100 linewidths and can be varied continuously.

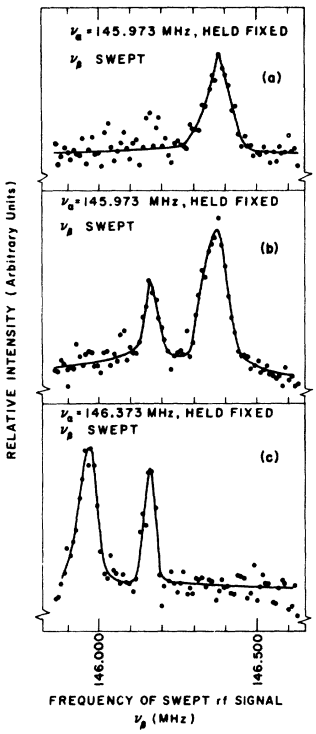

FIG. 2. Results of rf sweeps in the vicinity of 146.173 MHz, the frequency required for the normal two-quantum transition. A second fixed-frequency signal was applied at 145.973 MHz for (a) and (b), and at 146.373 MHz for (c). Curve (b) differs from (a) only in the use of higher rf power for (b). The two-frequency resonance at the right in (a) and (b) and at the left in (c) is broader than the normal two-quantum transition, and the frequency at which it occurs is continuously adjustable and determined by the choice of the fixed-frequency signal.

of the two frequencies ν_α and ν_β to the same rf loop. The $\Delta M_F = \pm 2$ transition indicated at the left in Fig. 1(b) was observed to be strong at $\nu_\beta \approx \nu_2$ when $\nu_\alpha = \nu_1$, as expected. It was also found, however, that a resonance could be observed for *any* value of ν_α within ±4 MHz of ν_1 if ν_β were swept through the appropriate range. Over the entire 8-MHz range of ν_α investigated, the requirement for resonance was found to be that the second signal have the frequency

$$\nu_\beta = (\nu_1 + \nu_2) - \nu_\alpha = [292.346(11)\ \text{MHz}] - \nu_\alpha ,$$

as expected for a two-quantum transition. For the particular power levels used, no frequency pulling was observed to within the experimental uncertainty of ±0.017 MHz ($\frac{1}{4}$ of Γ, the full width at half-maximum).

Figure 2 shows several of the two-frequency spectra obtained. For each curve, ν_α was held fixed as noted on the figure and ν_β was swept as shown. For the particular spectra displayed, both ν_α and ν_β are about 4 MHz (more than 50 linewidths) from the single-quantum resonance frequencies ν_1 and ν_2; and even with relatively high rf power the resonance is weak, as indicated by the poor signal-to-noise ratio. (The signal becomes much stronger if ν_α and ν_β are chosen closer to the single-quantum intervals ν_1 and ν_2.) Although spectrum (a) of Fig. 2 shows the two-quantum transition for which the quanta are unequal, it fails to show a resonance at $\nu_{2Q} = 146.173(6)$ MHz, the frequency required for the normal two-quantum transition. An attempt was then made to induce it by sweeping ν_β at a still higher rf power. Spectrum (b) of Fig. 2 is the result, and the normal double-quantum transition can now be seen at the proper frequency. (It is interesting to note that for the rf power levels used, the two-quantum transition with $\nu_\alpha \neq \nu_\beta$ has a greater transition probability than the normal two-quantum transition.) A repetition of this sweep, except that ν_α was turned off, showed the normal two-quantum transition as in (b), but the $\nu_\alpha \neq \nu_\beta$ two-quantum transition at the right in spectra (a) and (b) of Fig. 2 disappeared as expected. For spectrum (c), the fixed frequency ν_α was held at the frequency of the $\nu_\alpha \neq \nu_\beta$ resonance observed in spectra (a) and (b), and ν_β was again swept. As can be seen, the $\nu_\alpha \neq \nu_\beta$ two-quantum transition has now moved to the other side of the normal two-quantum transition in order to keep $\nu_\alpha + \nu_\beta = \nu_1 + \nu_2$, as required by energy conservation. (It can be seen from Fig. 2 that the width of the normal two-quantum transition is considerably less than that for the case in which the two frequencies are different, as predicted by Hack.[6])

It should be noted that none of the values of ν_α and ν_β for which the resonance was observed are consistent with the known properties of the Sn^{119} atom (except for the special case in which $\nu_\alpha \approx \nu_1$ and $\nu_\beta \approx \nu_2$); only the sum $\nu_\alpha + \nu_\beta$ contains any information about the structure of the atom.

III. CONCLUSIONS

Let us now consider how best to interpret observations of $\Delta F = 0$ resonance frequencies made at larger and larger fields by means of the two-frequency technique. For each transition, we will have a series of values of ν_α, ν_β, and the field H at which they were measured. If no $\Delta F = \pm 1$ intervals are known, A and B (and possibly g_J and g_I as well) are known only crudely and hence the $\Delta F = 0$, $\Delta M_F = \pm 1$ resonance frequencies are poorly known. Consequently, it may not be clear whether the observed resonance is due to two single-quantum jumps or to one two-quantum jump. Without further information, we cannot put ν_α and ν_β into the frequency-fitting routine individually; but the sum $\nu_\alpha + \nu_\beta$ can be put into the routine and will certainly be useful in reducing the uncertainty in the hfs constants and g_J value.

One can do more, however. One could change ν_α enough to move decisively off the $\Delta M_F = \pm 1$ energy interval (e.g., increase ν_α by say 5Γ, where Γ is the observed linewidth) and then repeat the run. If the observed frequency ν_β for resonance moves about 5Γ in the opposite direction, this fact would indicate that the resonance is indeed a two-quantum transition. If the resonance disappears,

the interpretation is unfortunately ambiguous since it may show either that single-quantum transitions were involved (and that ν_α and ν_β are individually meaningful) or that the run has failed for some other reason. Runs with ν_α changed by an amount less than Γ were found useful by Prior for identification of his observed resonances.

The two-frequency transitions seen by Prior *et al.*[3] evidently were due to successive single-quantum transitions since the observed frequencies for resonance not only permitted them to predict and observe the $\Delta F = \pm 1$ transitions, but all the data (both $\Delta F = 0$ and $\Delta F = \pm 1$) were self-consistent. The likelihood that two-quantum transitions for which $\nu_\alpha \neq \nu_\beta$ would be mistakenly identified as due to successive single-quantum transitions is probably smaller for radioactive detection than for stable-isotope work with a universal detector.

Although resonances due to two-quantum transitions may be misinterpreted as arising from successive single-quantum transitions, they may nevertheless be more useful in some cases than the latter as a spectroscopic tool. If the hyperfine interaction constants A and B and the g values g_J and g_I are not known well enough that frequencies analogous to ν_1 and ν_2 at a particular value of the field can be predicted to within a linewidth, the technique of successive single-quantum transitions is very difficult to use since *both* applied frequencies must be near resonance to induce a transition. In order to induce the two-quantum transition, however, it is only necessary to apply one frequency within ϵ of one $\Delta M_F = \pm 1$ resonant frequency, and to search a frequency interval of width 2ϵ around the other $\Delta M_F = \pm 1$ resonant frequency. The technique has been found useful[7] in investigation of the hfs of metastable atomic states of $Nd^{143,145}$. If a transition is observed, the sum of the two applied frequencies can be used to obtain quantitative information about the hyperfine structure.

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[1]Single-frequency magnetic-dipole multiple-quantum transitions have been observed by many authors. See, for example, P. Kusch, Phys. Rev. 93, 1022 (1954); R. L. Christensen, D. R. Hamilton, A. Lemonick, F. M. Pipkin, J. B. Reynolds, and H. H. Stroke, *ibid.* 101, 1389 (1956).

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[6]M. N. Hack, Ref. 4.

[7]W. J. Childs (unpublished).

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Comment on the Theory of Quantized Vortices*

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A recent statement that significant differences in the structure of a vortex line appear between the simple Hartree calculation and the variational Jastrow approach is shown to have no basis in reality.

In a recent paper,[1] Chester, Metz, and Reatto calculated the structure of vortices in He II using a Jastrow wave function whose form was determined variationally. One of the conclusions is that this calculation may suggest the limit of applicability of the simple Hartree theory[2] to this type of problem. "The density distribution in the core region is markedly different and the core size is somewhat smaller."[1] To substantiate this statement and in order to motivate *post facto* "The obvious objection to this kind of theory is that by its very nature it is unlikely to give an accurate description of phenomena which take place in spatial regions of the order of a few interatomic spacings,"[1] Figs. 1 and 2 are presented in Ref. 1.

We have reproduced these two figures below. In Fig. 1 we have added curve D to Fig. 1 of Ref. 1 to represent the energy versus the parameter a [Eq. (3.10) of Ref. 2]. Figure 1 shows that the difference in energy between the Hartree calculation and that of Chester *et al.* is even smaller than indicated by the dashed curve (which is a wrong representation