Proposed molecular test of local hidden-variables theories*

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The inefficiency of detectors of low-energy photons permits a loophole in the experimental refutation of local hidden-variables theories by observations of polarization correlations. A specific local hidden-variables model of Clauser and Horne shows that a hypothetical selectivity of detectors can yield the counting rates predicted by quantum mechanics. We therefore derive a new Bell-type inequality, which takes detection inefficiency into account. The loophole can then be blocked by observing spin correlations of atom pairs in an electronic singlet state, if both spin analysis and detection are sufficiently efficient. We propose to create Na atom pairs in the electronic singlet state, by using the induced Raman effect to excite Na_2 molecules in the electronic ground state $X\,^1\Sigma_g^+$ to the continuum of the electronic ground state. Two laser beams are to be used, one for excitation to a virtual intermediate state in $A\,^1\Sigma_u^+$, one to induce the downward transition to the final state. The ensemble needed to test the new inequality consists of those pairs which enter the respective collimating apertures of two Stern-Gerlach analyzers.

I. MOTIVATION

In 1935 Einstein, Podolsky, and Rosen[1] (EPR) considered a pair of correlated but spatially separated particles in hope of showing that the quantum-mechanical (QM) description must be supplemented by hidden variables. Bell[2] and his followers[3] proved the theorem that in certain cases the predictions of every local hidden-variables theory (LHVT) concerning such pairs disagree with those of QM. In principle, therefore, it is possible to perform experiments in order to test the entire family of LHVT against QM. A number of experiments have in fact been performed, and for the most part they have favored QM.[3] The experiments cannot be regarded as decisive, however, because of two loopholes. One of these loopholes is due to the theoretical possibility of communication, without action at a distance, between the parts of the experimental apparatus used to examine the spatially separated particles. We shall not be concerned with this loophole, but merely shall note that an experiment is being carried out by Aspect[4] for the purpose of blocking it. The other loophole arises from the fact that in each experiment performed so far, the analyzer-detector assemblies have been inefficient and only a small percentage of the ensemble of pairs of interest has been detected. Clauser and Horne[5] have constructed a model governed by a LHVT which nevertheless yields the same predictions as QM if the analyzer-detector assemblies are sufficiently inefficient. The purpose of this paper is to exhibit the feasibility of an experiment in which this loophole is blocked.

In order to deal conveniently with inefficiency, we shall follow the approach of Clauser and Horne to Bell's theorem, but with certain variations. The ensemble of interest will consist of pairs of particles entering the collimating apertures of two spatially separated analyzers (labeled "analyzer 1" and "analyzer 2"). Each analyzer has two output channels, labeled "+" and "−", and an incoming particle can be detected in at most one of these channels and need not be detected in either. It will be convenient to consider nondetection to be a third channel, labeled "0". Each analyzer has an adjustable parameter (such as an angle of orientation), a and a' being possible values of the parameter of analyzer 1, and b and b' being possible values of the parameter of analyzer 2. Let Λ be the space of complete states of particle pairs, with the possibility left open that there are hidden variables which give a state $\lambda \in \Lambda$ more content than any QM state. No assumption is made that the complete state λ of a particle pair determines the outcome of any experiment performed upon the pair, but it is assumed that the possible outcomes have definite probabilities fixed by λ and the conditions of the experiment. Specifically, there is a definite probability $P^{++}(a, b, \lambda)$ that particle 1 will be detected in channel + of analyzer 1 and particle 2 will be detected in channel + of analyzer 2, upon supposition that the parameters of the respective analyzers are a and b and the complete state of the particle pair is λ. $P^{+-}(a, b, \lambda)$, $P^{+0}(a, b, \lambda)$, etc., have analogous meanings. Clearly,

$$P^{++}(a,b,\lambda)+P^{+-}(a,b,\lambda)+P^{+0}(a,b,\lambda)+P^{-+}(a,b,\lambda)+P^{--}(a,b,\lambda)+P^{-0}(a,b,\lambda)+P^{0+}(a,b,\lambda)+P^{0-}(a,b,\lambda)+P^{00}(a,b,\lambda)=1, \tag{1}$$

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and each term on the left-hand side (lhs) of Eq. (1) is non-negative. The theory will be called *local* if

$$P^{++}(a,b,\lambda) = P_1^{+}(a,\lambda)P_2^{+}(b,\lambda), \tag{2a}$$

$$P^{+-}(a,b,\lambda) = P_1^{+}(a,\lambda)P_2^{-}(b,\lambda), \tag{2b}$$

$$P^{+0}(a,b,\lambda) = P_1^{+}(a,\lambda)P_2^{0}(b,\lambda), \tag{2c}$$

etc. (nine equations in all), where $P_1^{+}(a,\lambda)$ is the probability that particle 1 will be detected in channel + if the parameter of analyzer 1 is a and the state of the particle pair is λ, and $P_1^{-}(a,\lambda)$, $P_2^{+}(b,\lambda)$, and $P_2^{-}(b,\lambda)$ have similar meanings, and $P_1^{0}(a,\lambda)$ and $P_2^{0}(b,\lambda)$ are probabilites of nondetection. Now suppose that in this ensemble of particle pairs there is a probability distribution ρ over Λ. Then we define

$$P_1^{+}(a) = \int_\Lambda P_1^{+}(a,\lambda)d\rho, \tag{3a}$$

$$P_2^{+}(b) = \int_\Lambda P_2^{+}(b,\lambda)d\rho, \tag{3b}$$

$$P^{++}(a,b) = \int_\Lambda P^{++}(a,b,\lambda)d\rho, \tag{3c}$$

etc. An argument of Clauser and Horne leads to the following inequalities, regardless of the character of Λ and the choice of ρ:

$$P^{++}(a,a',b,b') \leq 0, \tag{4a}$$

$$P^{+-}(a,a',b,b') \leq 0, \tag{4b}$$

$$P^{-+}(a,a',b,b') \leq 0, \tag{4c}$$

$$P^{--}(a,a'b,b') \leq 0, \tag{4d}$$

where

$$P^{++}(a,a',b,b') \equiv P^{++}(a,b) - P^{++}(a,b') + P^{++}(a',b) + P^{++}(a',b') - P^{+}(a') - P^{+}(b), \tag{5}$$

and $P^{+-}(a,a',b,b')$, $P^{-+}(a,a',b,b')$, $P^{--}(a,a',b,b')$ have analogous meanings.

The QM predictions for certain choices of the ensemble of particle pairs and certain choices of a,a',b,b' violate these inequalities. In particular, the conclusion of three experiments performed with pairs of photons produced in certain atomic cascades violate inequality (4a) and confirm QM.[6-8] In these experiments photons were detected only in the + channel (which was taken to be passage through a linear polarizer), and the − channel (nonpassage) was lumped with the 0 channel. The photodetectors used in these experiments were only 10% to 30% efficient, so that relatively few of the photon pairs which passed through the respective polarizers were actually detected. Because of the inefficiency of the detectors it was not possible to infer the violation of inequality (4a) without making an additional (untestable) hypothesis[3]: Either (i) given that a pair of photons emerges from the polarizers, the probability of their joint detection is independent of the parameters a and b; or (ii) for every pair of photons from the source, the probability of a count with a polarizer in place is less than or equal to the corresponding probability with the polarizer removed. If neither of these assumptions is made, then a specific local hidden-variables model of Clauser and Horne[5] shows that a hypothetical selectivity of detectors can yield the counting rates predicted by QM. The need for an additional assumption shows that there is a loophole in previous experimental refutations of LHVT.

II. A NEW INEQUALITY

There is no need to invoke assumptions (i) or (ii) if the process of analysis and detection is sufficiently efficient, as we shall demonstrate. Consider beams of particles of the type under consideration entering the collimating apertures of the two analyzers (type being defined by particle species, approximate energy, and possibly other properties). Let $\eta_1(a)$ be the proportion of the particles entering the first analyzer which are detected in either the + or the − channel when the analyzer is oriented at angle a. Let $\eta_2(b)$ have an analogous meaning. We anticipate that in the proposed experiment it will be found that $\eta_1(a) = \eta_2(b) \equiv \eta$ for all angles a and b, but we shall first derive an inequality which is valid for any efficiencies. We are given, then,

$$P_1^{+}(a) + P_1^{-}(a) = \eta_1(a), \tag{6a}$$

$$P_2^{+}(b) + P_2^{-}(b) = \eta_2(b), \tag{6b}$$

but we cannot infer a definite probability that a pair of particles will be detected, because the detections of the two individual particles of the pair may be correlated in a manner unknown to us. Clearly, however, we can assert the inequality

$$1 - \min[\eta_1(a), \eta_2(b)] \leq P^{0+}(a,b) + P^{0-}(a,b) + P^{00}(a,b) + P^{+0}(a,b) + P^{-0}(a,b)$$

$$\leq P^{0+}(a,b) + P^{0-}(a,b) + P^{00}(a,b) + P^{+0}(a,b) + P^{-0}(a,b) + P^{00}(a,b)$$

$$= [1 - \eta_1(a)] + [1 - \eta_2(b)] = 2 - \eta_1(a) - \eta_2(b).$$

Hence

$$\eta_1(a)+\eta_2(b)-1\leq P^{++}(a,b)+P^{+-}(a,b)+P^{-+}(a,b)+P^{--}(a,b)\leq \min[\eta_1(a),\eta_2(b)]\,. \tag{7}$$

If we add inequality (4a) and inequality (4d) and use Eqs. (6a) and (6b) we obtain

$$P^{++}(a,b)+P^{--}(a,b)-P^{++}(a,b')-P^{--}(a,b')+P^{++}(a',b)+P^{--}(a',b)+P^{++}(a',b')+P^{--}(a',b')$$
$$\leq P_1^{+}(a')+P_1^{-}(a')+P_2^{+}(b)+P_2^{-}(b)=\eta_1(a')+\eta_2(b)\,. \tag{8}$$

Now we define the renormalized probabilities $\bar{P}^{++}(a,b)$, $\bar{P}^{+-}(a,b)$, etc., by

$$\bar{P}^{++}(a,b)\equiv\frac{P^{++}(a,b)}{P^{++}(a,b)+P^{+-}(a,b)+P^{-+}(a,b)+P^{--}(a,b)}\,. \tag{9}$$

$\bar{P}^{++}(a,b)$ is the probability that a pair will be detected in the + channels of both analyzers, conditional upon its being detected at all, and similarly for $\bar{P}^{+-}(a,b)$, etc. Then inequalities (7) and (8) yield

$$[\eta_1(a)+\eta_2(b)-1][\bar{P}^{++}(a,b)+\bar{P}^{--}(a,b)]-\min[\eta_1(a),\eta_2(b')][\bar{P}^{++}(a,b')+\bar{P}^{--}(a,b')]$$
$$+[\eta_1(a')+\eta_2(b)-1][\bar{P}^{++}(a',b)+\bar{P}^{--}(a',b)]+[\eta_1(a')+\eta_2(b')-1][\bar{P}^{++}(a',b')+\bar{P}^{--}(a',b')]$$
$$\leq\eta_1(a')+\eta_2(b)\,. \tag{10}$$

Now we shall consider the important special case

$$\eta_1(a)=\eta_1(a')=\eta_2(b)=\eta_2(b')\equiv\eta>\tfrac{1}{2}\,. \tag{11}$$

Inequality (10) becomes

$$S(a,a',b,b')\equiv\bar{P}^{++}(a,b)+\bar{P}^{--}(a,b)-[\eta/(2\eta-1)][\bar{P}^{++}(a,b')+\bar{P}^{--}(a,b')]$$
$$+\bar{P}^{++}(a',b)+\bar{P}^{--}(a',b)+\bar{P}^{++}(a',b')+\bar{P}^{--}(a',b')$$
$$\leq 2\eta/(2\eta-1)\,. \tag{12}$$

For simplicity of exposition in comparing the predictions of QM with those of LHVT we shall assume that Eq. (11) is correct. It will be seen in the following section that the predictions of QM disagree with inequality (12) in a specific experimental situation, provided that $\eta>0.9$.

It shall finally be noted that the renormalized probabilities $\bar{P}^{++}(a,b)$, etc., can be related easily to experimental quantities. If $R^{++}(a,b)$ is the rate at which pairs form the ensemble of interest are detected in coincidence in the + channels of both analyzers, and $R^{+-}(a,b)$, $R^{-+}(a,b)$, and $R^{--}(a,b)$ have analogous meanings, then

$$\bar{P}^{++}(a,b)\equiv\frac{R^{++}(a,b)}{R^{++}(a,b)+R^{+-}(a,b)+R^{-+}(a,b)+R^{--}(a,b)}\,. \tag{13}$$

There remains the problem of eliminating the accidental coincident counts due to unpaired particles and due to noise, so as to determine $R^{++}(a,b)$, etc., correctly. The method of doing this will be sketched in Sec. VI.

III. QUANTUM-MECHANICAL PREDICTIONS OF DETECTION PROBABILITIES

QM describes a pair of spin-$\frac{1}{2}$ particles in the singlet spin state by the vector

$$\Psi=[U_{\hat{n}}^{+}(1)\otimes U_{\hat{n}}^{-}(2)-U_{\hat{n}}^{-}(1)\otimes U_{\hat{n}}^{+}(2)]/(2)^{1/2}\,. \tag{14}$$

To simplify the initial discussion, suppose that particles 1 and 2 are propagating along $\hat{y}$ and $-\hat{y}$, respectively (a supposition which will later be removed because of experimental constraints). We can then choose the orientations $\hat{a}$ and $\hat{b}$ of the respective Stern-Gerlach magnetic fields to be in the plane perpendicular to $\hat{y}$, and a and b are the angles from $\hat{z}$ to $\hat{a}$ and $\hat{b}$, respectively, both measured in the counter-clockwise direction with respect to $\hat{y}$. Then the QM probability for detection of particle 1 in the + (−) channel of a Stern-Gerlach spin analyzer with magnet axis oriented along $\hat{a}$ is

$$[P_1^{\pm}(a)]_{\Psi}=\eta/2\,, \tag{15a}$$

and similarly

$$[P_2^{\pm}(b)]_{\Psi}=\eta/2\,, \tag{15b}$$

where Eq. (11) has been assumed. Moreover, the QM probabilities for joint detection in the designated channels are

$$[P^{++}(a,b)]_{\Psi}=[P^{--}(a,b)]_{\Psi}=\eta^2[1-\cos(a-b)]/4\,, \tag{16a}$$

$$[P^{+-}(a,b)]_{\Psi}=[P^{-+}(a,b)]_{\Psi}=\eta^2[1+\cos(a-b)]/4\,. \tag{16b}$$

The efficiency η of a single analyzer-detector assembly shows up in the right-hand side (rhs) of Eqs. (15a) and (15b). In the usual QM analysis, the efficiency of the pair of analyzer-detectors is η^2, as exhibited in the rhs of Eqs. (16a) and (16b). (As we pointed out in Sec. II, we were not entitled to make this assumption in a general LHVT.)

In the experiment which we are proposing, the particle pairs will be Na atoms obtained by dissociation (within a small interaction region about the origin O) of a beam of Na_2 molecules incident along $-\hat{x}$, as shown in Fig. 1. There will be a distribution over pairs of directions of the daughter atoms, which is peaked at the directions $\hat{y}'$ and $\hat{y}''$ indicated in Fig. 1. The analyzer-detector assemblies are located so that the line from O to the center of the collimating aperture of the first assembly lies along $\hat{y}'$, while that from O to the center of the collimating aperture of the second assembly lies along $\hat{y}''$. $\hat{y}'$ and $\hat{y}''$ are obtained by counter-clockwise rotations of $\hat{y}$ about $\hat{z}$ through the angles θ and $\pi - \theta$, respectively. Suppose that initially the two Stern-Gerlach fields are oriented along $\hat{z}$. Then a is the angle by which the first field is rotated counter-clockwise about $\hat{y}'$, and b is the angle by which the second field is rotated counter-clockwise about $\hat{y}''$. Notice that the interpretations of a and b are different from the interpretations in the simple case discussed at the beginning of this section, where the counter-clockwise rotations were both with respect to the positive y axis. With the new meanings of a and b,

$$[P^{++}(a,b)]_\Psi = \langle\Psi\,|E_1(\theta,a,0)\otimes E_2(\pi-\theta,b,0)\,|\Psi\rangle\,, \quad (17a)$$

where $E_1(\theta,a,0)$ is the projection in the spin space of atom 1 onto the state obtained by the rotation $(\theta,a,0)$ from a state of spin up along $\hat{z}$; and $E_2(\pi-\theta,b,0)$ has an analogous meaning. Likewise,

$$[P^{+-}(a,b)]_\Psi = \langle\Psi\,|E_1(\theta,a,0)\otimes[1-E_2(\pi-\theta,b,0)]\,|\Psi\rangle\,, \quad (17b)$$

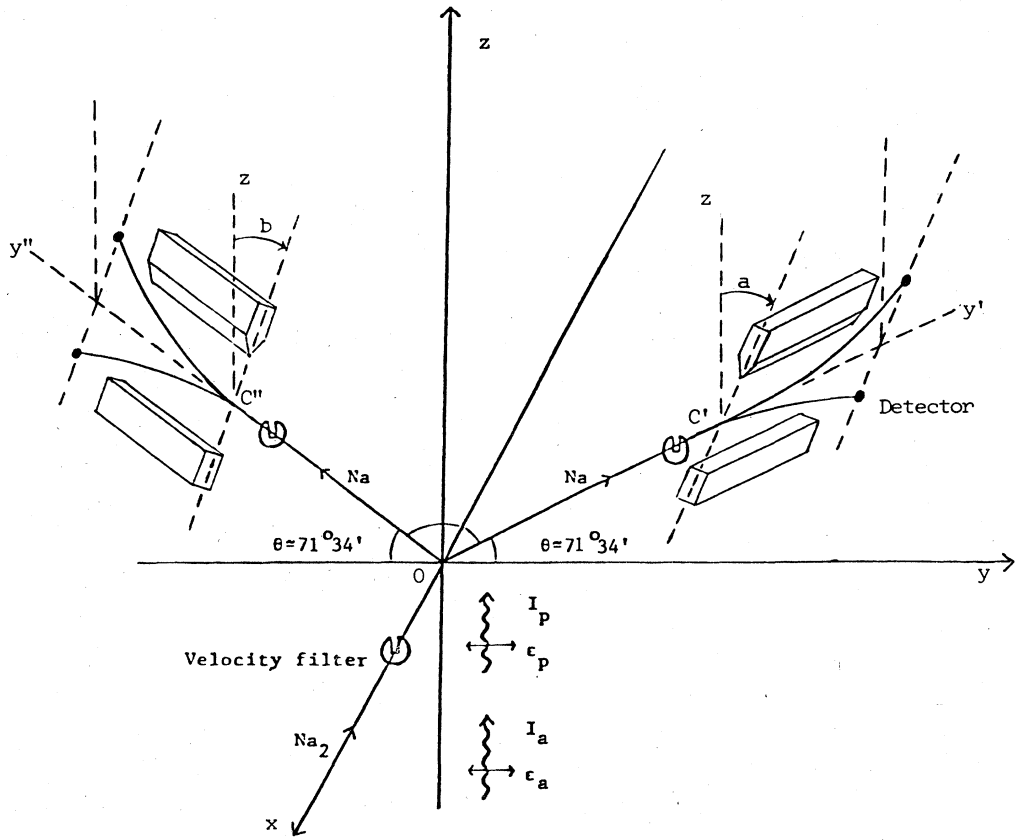

FIG. 1. Proposed experimental arrangement. Incident beam is along $-\hat{x}$. Polarizations of pumping and auxiliary beams are along the y axis. C' is center of aperture of assembly (1). C'' is center of aperture of assembly (2). Trajectory OC' of first daughter atom is along $\hat{y}'$. Trajectory OC'' of second daughter atom is along $\hat{y}''$. a and b are angles of orientations of the respective Stern-Gerlach fields.

$$[P^{-+}(a,b)]_\Psi = \langle\Psi|[1-E_1(\theta,a,0)]\otimes E_2(\pi-\theta,b,0)|\Psi\rangle\,, \tag{17c}$$

$$[P^{--}(a,b)]_\Psi = \langle\Psi|[1-E_1(\theta,a,0)]\otimes[1-E_2(\pi-\theta,b,0)]|\Psi\rangle\,. \tag{17d}$$

In computing $[P^{++}(a,b)]_\Psi$, etc., it is convenient to take $\hat{n}$ in Eq. (14) to be $\hat{z}$, which is justified by the spherical symmetry of Eq. (14). Then,

$$[P^{++}(a,b)]_\Psi = [P^{--}(a,b)]_\Psi$$
$$= \eta^2[\cos^2(a/2)\sin^2(b/2)+\sin^2(a/2)\cos^2(b/2)+2\cos(a/2)\sin(a/2)\cos(b/2)\sin(b/2)\cos(2\theta)]/2\,, \tag{18a}$$

$$[P^{+-}(a,b)]_\Psi = [P^{-+}(a,b)]_\Psi$$
$$= \eta^2[\cos^2(a/2)\cos^2(b/2)+\sin^2(a/2)\sin^2(b/2)-2\cos(a/2)\sin(a/2)\cos(b/2)\sin(b/2)\cos(2\theta)]/2\,. \tag{18b}$$

The renormalized probabilities are

$$[\bar{P}^{++}(a,b)]_\Psi \frac{[P^{++}(a,b)]_\Psi}{[P^{++}(a,b)]_\Psi+[P^{+-}(a,b)]_\Psi+[P^{-+}(a,b)]_\Psi+[P^{--}(a,b)]_\Psi} = (1/\eta^2)[P^{++}(a,b)]_\Psi\,, \tag{19}$$

and similarly for $[\bar{P}^{+-}(a,b)]_\Psi$, $[\bar{P}^{-+}(a,b)]_\Psi$, and $[\bar{P}^{--}(a,b)]_\Psi$. (In other words, the efficiency factor η^2 on the rhs of Eqs. (18a) and (18b) is canceled out.)

The angle θ is defined in terms of the ratio of the mean velocity of Na_2 molecules parallel to the molecular beam and the dissociation velocity of the Na atoms resulting from the dissociative excitation process. In the experiment proposed in Sec. IV,

$$\theta = \tan^{-1}(3) \cong 71°34'\,. \tag{20}$$

Modifications of the details of our proposal would, of course, change θ. For the θ of Eq. (20), a convenient choice of the angles [which has the effect of maximizing the deviation of the QM prediction from inequality (12) when $\eta = 1$] is

$$a = 270°,\quad a' = 0°\quad b = 141°20',\quad b' = 218°40'\,. \tag{21}$$

With these angles

$$[S(a,a',b,b')]_\Psi \equiv [\bar{P}^{++}(a,b)]_\Psi + [\bar{P}^{--}(a,b)]_\Psi - [\eta/(2\eta-1)]\{[\bar{P}^{++}(a,b')]_\Psi + [\bar{P}^{--}(a,b')]_\Psi\}$$
$$+ [\bar{P}^{++}(a',b)]_\Psi + [\bar{P}^{--}(a',b)]_\Psi + [\bar{P}^{++}(a',b')]_\Psi + [\bar{P}^{--}(a',b')]_\Psi$$
$$= 0.3749 + 0.3749 - [\eta/(2\eta-1)](0.1250+0.1250+0.4452+0.4452+0.4452+0.4452$$
$$= 2.5306 - 0.25[\eta/(2\eta-1)]\,. \tag{22}$$

Equation (22) disagrees with inequality (12) provided that

$$\eta > 0.9 \tag{23}$$

Consequently, if the Na atom detectors are efficient enough, one can test the family of LHVT against QM without using the additional assumptions mentioned in Sec. I.

IV. PROPOSED EXPERIMENTAL ARRANGEMENT

Figure 1 is a schematic diagram of the proposed experiment. The Na_2 molecular beam is produced by expansion of Na vapor through a nozzle. The supersonic molecular beam passes through a velocity filter to assure a narrow velocity distribution for the molecules. When the molecular beam reaches the interaction region, it is intercepted by two laser beams I_b, I_a, the "pumping" beam, and the auxiliary "inducing" beam. The linear polarizations of the laser beams are chosen to be along the y axis. If the molecular velocities are not taken into account, the spatial distribution of Na atoms from dissociation of Na_2 molecules peaks about the y axis. With molecular velocities taken into account, the actual spatial distribution of Na atoms peaks in the xy plane and along directions defined by the angle θ. Two Stern-Gerlach magnets are placed along these two directions. Each Na atom of the atom pair goes through a velocity filter and then enters the Stern-Gerlach magnet. The gradient of the inhomogeneous magnetic field is great enough that Na atoms with electronic spin up along the field direction are well separated from those with electronic spin down. Finally, a Na atom from a pair is detected by a surface ionizer in coincidence with the other atom from the same pair.

Figure 2 indicates the dissociative excitation process of the induced Raman effect. $X^1\Sigma_g^+$ is the

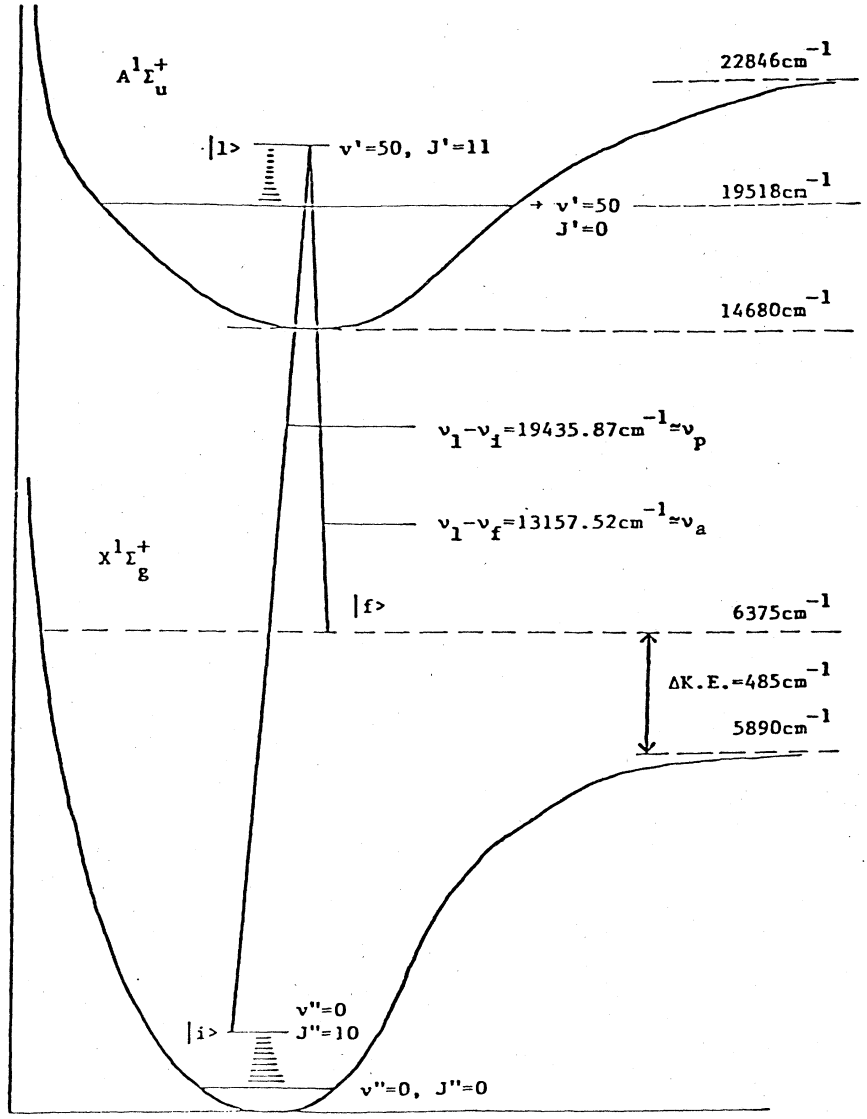

FIG. 2. Proposed laser-induced transition in Na_2. $|i>$, $|l>$, and $|f>$ are respectively the initial, intermediate, and final states of Na_2 in the proposed transition. (References 9–11.)

electronic ground state and $A^1\Sigma_u^+$ is the electronic first excited state.[9-11] Both states have zero total electronic spin. The pumping beam I_p is tuned close to resonance with the particular chosen transition frequency between the initial state in the electronic ground state $X^1\Sigma_g^+$ and the intermediate state in the electronic excited state $A^1\Sigma_u^+$. The auxiliary inducing beam I_a is used to induce the downward transition to the final state in the vibrational continuum of the ground electronic state. Since the Na_2 molecule dissociates in the continuum region of the electronic ground state, the electronic spins of the newly created Na atom pair form the required electronic singlet state.

V. RATE OF PRODUCTION OF MEMBERS OF THE ENSEMBLE

We wish to make a numerical estimate of the rate R_0 at which both of the Na atoms resulting from the dissociation of a Na_2 molecule enter the collimating apertures of the two Stern-Gerlach magnets, since it is these pairs that constitute the ensemble of interest. In order to calculate R_0 we shall make tentative choices of the initial, intermediate, and final molecular states, and shall use parameters of the beams and apparatus which are found in the experimental literature.

The molecular states involved in the induced

Raman effect will be the initial state $|i\rangle = |X^1\Sigma_g^+, v''=0, J''=10\rangle$, the intermediate state $|l\rangle = |A^1\Sigma_u^+, v'=50, J'=11\rangle$, and the final state $|f\rangle = |X^1\Sigma_g^+$, 485 cm^{-1} above the dissociation limit$\rangle$. The molecular states are chosen such that: (i) the population of Na_2 molecules is maximum in the chosen initial state, (ii) the product of the two Franck-Condon factors involving the vibrational levels is large, while at the same time the trajectories of the Na atoms of a pair deviate enough from each other to ensure easy Stern-Gerlach analysis of the atoms.

Using results of various studies on the characteristics of supersonic alkali beams,[12-16] we assume that the Na beam has the following characteristics: Total beam intensity $\cong 4\times10^{19}$ particles/sr sec; Na_2 concentration $\cong 20\%$; vibrational temperature $\cong 150°K$; rotational temperature $\cong 50°K$; the distribution of the velocity parallel to the beam axis for the molecules in the state $|i\rangle$ has the form

$$n(v) = nv^3 \exp[-S^2(v/u-1)^2],$$

where $u = 150\,000$ cm/sec and $S = u/(2kT/m)^{1/2} = 8.7$.

The Na_2 molecular beam originates 5 cm from the interaction region, where the collimated molecular beam has a cross section of $10^{-2}\times10^{-3}$ cm^2. The linearly polarized laser beams are focused to have cross sections of 10^{-6} cm^2 at the interaction region. Consequently, the interaction region is of dimension $10^{-2}\times10^{-6}$ cm^2. The small cross sections of the laser beams give large photon intensities, while the rectangular shape of the molecular beam is chosen in order to obtain a large number of molecules dissociating into pairs of atoms with trajectories appropriate for entering the collimating apertures. The pumping beam (from a cw tunable dye laser) is assumed to have power 0.1 W, wave number about 19 435 cm^{-1}, and line width about 100 MHz. It is tuned just off resonance with the transition frequency between the initial state $|i\rangle$ and the intermediate state $|l\rangle$. The auxiliary inducing beam is assumed to have power 1 W, wave number about 13 157 cm^{-1}, and line width of order 1 GHz. One of the existing laser lines[17] may suffice, so long as the deviation from the wave number 13 157 cm^{-1} does not produce great changes. The Stern-Gerlach magnets are placed 30 cm from the interaction region. They are similar to the ones used by Freeman, *et al.*[18] Their magnets are 25-cm-long, two-wire fields with maximum gradients of 34 kG/cm. The maximum usable beam deflection is about 0.2 cm. The velocity filters and the collimating apertures are 5 cm in front of the magnets, and the surface ionizers are 5 cm from the other ends of the magnets. The width of the ionizers is approximately 0.1 cm.[19]

The rate R_0 is given by the following expression:

$$R_0 = \frac{dp}{dt} Q\Delta T P(\mathrm{PAA})\delta^2 . \tag{24}$$

Here P(PAA) represents the P that the pair approaches apertures. The product of the first three terms gives the number of atom pairs produced per second by the induced Raman effect, and the product of the last two terms gives the probability that the two members of a Na atom pair will enter the respective collimating apertures of the Stern-Gerlach magnets. (dp/dt) is the QM transition probability for the induced Raman effect. It will be discussed below. δ^2 is an efficiency factor.

Q is the number of molecules going through the interaction region per second that are in the initial state $|i\rangle$. Q depends on the characteristics of the Na beam and is given by the following expression: Q = (total number of particles approaching interaction region per second) × (proportion of molecules among the particles) × (proportion of molecules in $v''=0$ level) × (proportion of molecules in $J''=10$ level) × (proportion of molecules having velocities within limits set by velocity filter) × (efficiency of velocity filter). The velocity filter has a range approximately ± 300 cm/sec from the mean molecular velocity, 150 000 cm/sec. The efficiency of the filter, which is the probability that a particle with velocity within the specified range will pass through the filter, is assumed to be 0.5, the upper limit for a thin disk-like filter.[20] Using the molecular beam characteristics mentioned earlier, we estimate Q to be about 1.3×10^9/sec. ΔT is the possible time for interactions between the molecules and the laser beams. It is simply the time for a molecule to travel through the interaction region. ΔT is approximately 7 nsec.

P(PAA) is the probability that two atoms of the same pair have the appropriate trajectories to approach their respective collimating apertures. The probability can be estimated by elementary but lengthy geometrical considerations.[20] With the apertures placed 25 cm from the interaction region and of approximate dimension 0.028×0.029 cm^2, P(PAA) is estimated to be 2.75×10^{-6}. δ is the efficiency of the velocity filters in front of the apertures. It is squared because the filtering processes of the two filters are essentially independent of each other.

The Raman effect is a second-order radiative process. The transition probability of the process can be calculated using the second-order contribution in time-dependent perturbation theory[21]:

$$\frac{dp}{dt} = \frac{2\pi}{\hbar^2}\left|\frac{1}{\hbar}\sum_L \frac{\langle F|H_{\rm int}|L\rangle\langle L|H_{\rm int}|I\rangle}{\omega_I - \omega_L}\right|^2 \delta(\omega_I - \omega_F),$$

where $|I\rangle$, $|L\rangle$, $|F\rangle$ are, respectively, the initial, intermediate, and final states of the molecule plus photon system; $E_I = \hbar\omega_I$ is the energy of the initial state, etc.

If the initial state of the molecule plus photon system is written as

$$|I\rangle = |n_p, n_a, i\rangle,$$

where n_p is the number of photons of pumping frequency ω_p, n_a is the number of photons of inducing frequency ω_a, and $|i\rangle$ is the molecular state; then the final and the intermediate states for the induced Raman effect are

$$|F\rangle = |n_p - 1, n_a + 1, f\rangle,$$

and

$$|L\rangle = |n_p - 1, n_a, l\rangle \quad \text{or} \quad |L\rangle = |n_p, n_a + 1, l\rangle.$$

Letting $e\vec{D}$ denote the dipole moment operator of the molecule and using the electric dipole (E1) approximation, we obtain the standard expression for the induced Raman effect:

$$dp/dt = 8\pi^3\alpha^2 n_p(n_a + 1)c^2V^{-2}\omega_p\omega_a \times\left|\sum_l [G(l) + H(l)]\right|^2 \delta(\omega_p - \omega_a + \omega_i - \omega_f),$$

with

$$G(1) = \langle f|\hat{\epsilon}_a\cdot\vec{D}|l\rangle\langle l|\hat{\epsilon}_p\cdot\vec{D}|i\rangle/(\omega_l - \omega_i - \omega_p),$$

$$H(1) = \langle f|\hat{\epsilon}_p\cdot\vec{D}|l\rangle\langle l|\hat{\epsilon}_a\cdot\vec{D}|i\rangle/(\omega_l - \omega_i + \omega_a),$$

where $\hat{\epsilon}_p$ and $\hat{\epsilon}_a$ are the polarization vectors for the pumping and auxiliary inducing beams, and ω_i, ω_l, and ω_f are frequencies corresponding to the energy levels of the initial, intermediate, and final states of the molecule.

In the proposed experiment the pumping frequency is tuned close to resonance with the transition frequency between the initial molecular state $|i\rangle$ and the intermediate state $|l\rangle$. As a result, the sum $\sum_l[G(l)+H(l)]$ reduces to the contribution of that single intermediate state

$$\frac{dp}{dt} = 8\pi^3\alpha^2 n_p(n_a + 1)c^2V^{-2}\omega_p\omega_a \times\left|\sum_{M'} G(l)\right|^2 \delta(\omega_p - \omega_a + \omega_i - \omega_f).$$

(We still have to sum over the magnetic quantum number M' of the state.)

Using the axial-recoil approximation[22] and performing the standard QM calculations, we obtain the final expression for the transition probability of the induced Raman effect[20]

$$\frac{dp}{dt} = 4\pi^2\alpha^2 c^{-1}B(J'')\left|d_{n'n''}\right|^4 q_{vv'}q_{v'v''} \times I_p I_a \nu_p \nu_a(\nu_l - \nu_i - \nu_p)^{-2}, \tag{25}$$

where

$$B(J'') = (J'' + 1)(4J'' + 3)/[15(2J'' + 1)^2] = 0.0715,$$

for $J'' = 10$; $d_{n'n''}$ is the average electronic transition moment (with factor of e omitted) between $X^1\Sigma_g^+$ and $A^1\Sigma_u^+$, and numerically it is about 2×10^{-8} cm[23];

$$q_{v'v''} = 3.553 \times 10^{-9},$$

$$q_{vv'} = 1.391 \times 10^{-3}\ \text{cm},$$

are the Franck-Condon factors[11,24];

$$I_p = 3 \times 10^{23}/\text{cm}^2\,\text{sec},$$

$$I_a = 3 \times 10^{24}/\text{cm}^2\,\text{sec},$$

are the photon number intensities of the pumping beam and the auxiliary inducing beam at the interaction region;

$$\nu_p \cong 19\,435\ \text{cm}^{-1},$$

$$\nu_a \cong 13\,157\ \text{cm}^{-1},$$

are the approximate wave numbers of the pumping beam and the auxiliary inducing beam;

$$(\nu_l - \nu_i - \nu_p) \cong 0.0033\ \text{cm}^{-1}$$

(equivalent to 100 MHz) is the difference between the pumping frequency and the transition frequency connecting the initial and the intermediate states of the molecule. The natural width of the intermediate state is not included in the factor because the radiative lifetime is about 12 nsec, which is only about 15 MHz.[25] Also the Doppler width is ignored because the molecular velocity parallel to the laser beams is quite small.

Using the above numerical values, we estimate R_0, the rate at which pairs of the ensemble of interest are produced, to be about 0.5 pairs/sec.

VI. DISCUSSION: DETECTION RATES AND EFFICIENCIES

In obtaining the experimental values of coincident detection rates of Na atom pairs, we need to consider the effect of accidental coincidences of unpaired Na atoms, where an unpaired atom is one which comes from a dissociating Na_2 molecule and enters a collimating aperture while its partner fails to enter the other aperture. (Accidental coincidences involving paired Na atoms from two distinct Na_2 molecules are negligible.) Also background noise in the detection process, which consists mainly of the evaporation of ions from the surface ionizers without the stimulus of incident

atoms, can give rise to accidental coincidences. To correct for accidental coincidences we proceed in two steps. We first check the rates $R_1^+(a)$, $R_1^-(a)$, $R_2^+(b)$, and $R_2^-(b)$ of single detections by the four detectors (when the orientations are a and b). According to QM, the rates should be equal for all orientations of the Stern-Gerlach magnets, if the analyzer-detector assemblies are equivalent and are symmetrically placed. Second, we vary the orientations of the two Stern-Gerlach magnets and determine the minimum coincident detection rates $R_{\min}^{++}$, $R_{\min}^{+-}$, $R_{\min}^{-+}$, and $R_{\min}^{--}$ for the four pairs of detectors. According to Eqs. (18a) and (18b), the contribution of coincidences from two atoms of a single pair to $R_{\min}^{++}$ and $R_{\min}^{--}$ is 0 when the orientations are parallel (i.e., $a=0$, $b=0$), and the contribution to $R_{\min}^{+-}$ and $R_{\min}^{-+}$ is 0 when the orientations are antiparallel. Furthermore, because the single detection rate is the same for each detector, the accidental coincident detection rates should be the same for all four pairs of detectors and for all orientations. Hence within experimental error, we should find

$$R_{\min}^{++}=R_{\min}^{+-}=R_{\min}^{-+}=R_{\min}^{--}=R_{\min} \ , \tag{26}$$

and the common value $R_{\min}$ is the accidental coincident detection rate. If Eq. (26) is not satisfied, there is either a discrepancy with QM or a systematic error, which must be investigated further. If, however, Eq. (26) is satisfied, then "corrected" detection rates are determined by subtracting $R_{\min}$ from the experimental rates, i.e.,

$$R^{++}(a,b)=R_{\text{expt}}^{++}(a,b)-R_{\min} \ , \tag{27a}$$

$$R^{+-}(a,b)=R_{\text{expt}}^{+-}(a,b)-R_{\min} \ , \tag{27b}$$

$$R^{-+}(a,b)=R_{\text{expt}}^{-+}(a,b)-R_{\min} \ , \tag{27c}$$

$$R^{--}(a,b)=R_{\text{expt}}^{--}(a,b)-R_{\min} \ . \tag{27d}$$

The corrected detection rates are to be used in checking inequality (12).

It remains to discuss the experimental determination of the efficiencies $\eta_1(a)$, $\eta_1(a')$, $\eta_2(b)$, and $\eta_2(b')$. As we noted in Sec. II, a comparison between the predictions of QM and those of LHVT can be made by means of inequality (10), even if these efficiencies are not all equal; but if they all have the same value η, we are able to use the simpler inequality (12). Furthermore, as we saw in Sec. III, QM conflicts with inequality (12) in the proposed experiment only if η is greater than 0.9.

To measure the efficiency $\eta_1(a)$ we need only compare $[R_1^+(a)+R_1^-(a)]$ with the actual rate at which Na atoms enter the first analyzer-detector assembly; and similarly in order to measure $\eta_1(a')$, $\eta_2(b)$, and $\eta_2(b')$. A suitable procedure for determining the actual entrance rate would be to maintain the experimental arrangement of Sec. IV with one modification: a collector plate is placed is placed between the collimating aperture and the Stern-Gerlach magnet. Na atoms will be deposited on the collector plate during a known time interval, and afterwards can be counted by the remarkably efficient "resonance ionization spectroscopy" technique discussed in a number of articles.[26,27]

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