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Nuclear acoustic resonance in single-crystal gold metal

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Nuclear acoustic resonance is reported for the first time for ^{197}Au in single-crystal gold metal. The ratio $\omega^{(197)}/H$ was measured to be $(2\pi)(74.118 \pm 0.007)$ Hz/G and the line shape of the $\Delta m = \pm 2$ resonance was a Lorentz line shape with a half width at half amplitude of 154.8 ± 1 G at 78 K after high-temperature annealing. $\vec{\vec{S}}$ tensor components which relate dynamic electric field gradients with dynamic elastic strain are determined to be of opposite sign and their magnitudes are $S_{11}-S_{12}=13.6\times10^{15}$ statvolts/cm^2 and $S_{44}=13.0\times10^{15}$ statvolts/cm^2. Lattice contributions to these components are computed and the conduction-electron contributions determined.

I. INTRODUCTION

Nuclear acoustic resonance (NAR) can often be used to study nuclear-spin systems in single-crystal metals in which the magnetic moment is very small [making nuclear magnetic resonance (NMR) difficult] but where there is a sizable quadrupole moment and large Sternheimer antishielding factor. In such cases, the NAR coupling between the high-frequency elastic wave and the nuclear-spin system can be chosen to be of nuclear electric quadrupole origin.

At a nuclear position in a crystal with cubic symmetry, a dynamic elastic strain will create a dynamic electric field gradient (DEFG). The DEFG relationship to the dynamic elastic strain can be written in terms of a tensor of the fourth rank, $\vec{\vec{S}}$. From crystal symmetry, there are three different nonzero components of $\vec{\vec{S}}$. When expressed in the Voigt notation, these are S_{11}, S_{12}, and S_{44}. Harrison and Sagalyn[1] have shown that the DEFG can always be written as a combination of the two independent tensor components $S_{11}-S_{12}$ and S_{44}. With NAR it is possible to make absolute measurements of the magnitudes and relative signs of $S_{11}-S_{12}$ and S_{44}.

In the only previous (NMR) study of ^{197}Au in gold metal, Narath[2] reported measurements of the spin-lattice relaxation time, the resonance center position, and estimated the frequency width of the $\frac{1}{2}$ to $\frac{1}{2}$ transition, all at 1.6 K.

In this paper we report the first NAR study of ^{197}Au in single-crystal gold. Measurements were made of the resonance line-center position, the resonance line shape and linewidth, and the magnitudes and relative signs of the tensor components $S_{11}-S_{12}$ and S_{44}.

II. EXPERIMENTAL DETAILS

A gold single crystal, spark-erosion cut into a right cylinder of length 0.95 cm and diameter 0.95 cm with cylindrical axis in the [110], 99.999% pure, was obtained from Aremco Products. The crystal was ground, polished, and etched to give a final specimen length of 0.902 cm. End faces of the cylinder were made flat and parallel to within 1.3 μm, and were measured by x-ray diffraction techniques to be within $\pm 0.1°$ of (110) crystal planes. The crystal was then annealed at 1030 °C for 16 h.

Acoustic waves were generated by means of X-cut (longitudinal) quartz piezoelectric transducers bonded to the specimen with Nonaq stopcock grease. The coaxial transducers were 0.953 cm in diameter with a center radiating area of 0.64 cm. NAR absorption measurments and Alpher-Rubin calibration measurements[3] were made at 10 MHz, with utilization of a standard marginal oscillator ultrasonic spectrometer.[4] The magnetic field of 7.3 T was supplied by an Oxford superconducting magnet with a 650-G sweep window.

The technique of NAR measures the acoustic attenuation change associated with the absorption of resonant acoustic energy by a nuclear-spin system.[4] The dominant coupling between elastic strains and the ^{197}Au nuclear-spin system is the dynamic nuclear quadrupole interaction. Longitudinal acoustic waves were used with propagation vector $\vec{k}$ along [110] and polarization vector $\vec{\epsilon}$ also along [110] and measurements were made as a function of the angle ξ which is the angle in the $(\bar{1}10)$ plane between the external magnetic field and the propagation direction. Using the notation of an earlier paper,[5] we write the amplitude of the dynamic attenuation change as

$$\Delta\alpha_{(\Delta m=\pm 2)} = CBQ^2[(-\cos^2\xi)\tfrac{1}{2}(S_{11}-S_{12}) + (1+\sin^2\xi)S_{44}]^2$$

and

$$B = \frac{\sum_m (I\mp m)(I\mp m-1)(I\pm m+1)(I\pm m+2)}{(2I)^2(2I+1)(2I-1)^2} ,$$

$$C = (\pi/64)Ne^2\omega^2 g(H)/(kT\rho v^3\gamma) ,$$

where I is the nuclear spin, N is the number of resonant nuclear spins per unit volume, ω is the resonant acoustic wave frequency, $g(H)$ is the normalized line-shape function in magnetic field units, ρ is the mass density, v is the acoustic wave velocity, k is the Boltzmann constant, T is the absolute temperature, Q the quadrupole moment, and γ the nuclear gyromagnetic ratio for $\Delta m = \pm 1$ transitions.

The ^{197}Au NAR was observed at 300 and 78 K. At 78 K, although saturation effects were observed, the acoustic power level could be reduced so that the saturation was less than 1%. Data were taken at 78 K using a modulation amplitude that was approximately 10% of the resonance width at half amplitude. Because of the approximate 300-G width at half amplitude of the absorption signal, five overlapping sweep windows were used at $\xi = 90°$ to record the NAR signal. For each window, the spectrometer gain was calibrated with the Alpher-Rubin calibration signal and the digitized

data channel numbers were calibrated with the $Al^{123}Sb$ NMR. The data from the five sweep windows were added together digitally. During these data-taking runs, the NAR frequency was stable to better than 2.5×10^{-5} and the magnetic field center was stable to better than 1×10^{-8}. From these data, the line shape, linewidth, and S_{44} were determined.

Similar measurements were made with a window centered around the ^{197}Au NAR at the angles $\xi=70°$ and 50° which allow determination of relative signs of $S_{11}-S_{12}$ and S_{44} and the magnitude of $S_{11}-S_{12}$.

Absolute magnetic field at the ^{197}Au NAR resonance center was determined at 78 K by raising the NAR sample above the magnet center and allowing an NMR coil containing single-crystal AlSb to be in the magnet center. The position of the $Al^{123}Sb$ NMR was then determined using the same magnetic field sweep parameters as used for the NAR data. The 78-K $Al^{123}Sb$ also was measured versus ^2H in D_2O at 300 K.

III. RESULTS AND DISCUSSION

The ^{197}Au experimental line shape when integrated digitally was an exact Lorentz line-shape function within experimental error with a half width at half maximum of 154.8 ± 1 G at 78 K after the high-temperature anneal. With successive temperature cycling between 78 and 300 K and transducer rebonding, the linewidth broadened while the integrated shape continued to be a Lorentz function. We compute that the square root of the theoretical second moment due to the dipole-dipole interaction is 0.0713 G for $\Delta m=\pm2$ transitions. The experimental line shape being a Lorentz function, the linewidth broadening under strain due to thermal cycling between 78 and 300 K and mechanical handling, and the very large ratio existing between the experimental width at half maximum and the computed dipole-dipole width suggest that inhomogeneous line broadening is present, possibly due to strain-induced electric field gradients.

In pure gold single crystal, the magnetic field was measured at 78 K at the center of the $\Delta m=\pm2$ NAR of ^{197}Au. The value of $\omega^{(197)}/H$ was found to be (2π) (74.118 ± 0.007) Hz/G. This value agrees within experimental error with the value found by Narath[2] of (2π) (74.125 ± 0.004) Hz/G at 1.6 K.

From the experimental angular dependence of the dynamic acoustic attenuation, the signs of QS_{44} and $Q(S_{11}-S_{12})$ are found to be different. Using the quadrupole moment[6] $Q=0.547\pm0.0016$ b, we find the experimentally determined $\overleftrightarrow{S}$ tensor components are $|S_{44}|=13.0\times10^{15}$ statvolts cm^{-2} (±4%) and $|S_{11}-S_{12}|=13.6\times10^{15}$ statvolts cm^{-2} (±10%), where the experimental error does not include the error in Q.

In order to further analyze the experimental $\overleftrightarrow{S}$ tensor components, we follow a suggestion by Watson, Gossard, and Yafet[7] and write

$$S_{\alpha\beta}=(1-\gamma_\infty)(S_{\alpha\beta})_{\text{latt}}+(S_{\alpha\beta})_{\text{ce}}\ ,$$

where $(S_{\alpha\beta})_{\text{latt}}$ is the nuclear plus the electronic contribution outside an atomic sphere centered around the nucleus in question, γ_∞ is the Sternheimer antishielding factor, and $(S_{\alpha\beta})_{\text{ce}}$ is the contribution of the conduction electrons inside the atomic sphere. Butet[8] has used the planewise summation method used first by De Wette[9] to compute for the fcc lattice,

$$S_{11\ \text{latt}}=10.293Zea^{-3}\ ,$$

where Ze is the charge of an ion and a is the gold lattice constant equal to 4.0781 Å. Butet shows that

$$(S_{11}-S_{12})_{\text{latt}}=\tfrac{3}{2}S_{11\ \text{latt}}$$

and

$$S_{44\ \text{latt}}=-\tfrac{1}{2}S_{11\ \text{latt}}\ .$$

By assuming a Z value of +1, we find, in units of statvolts cm^{-2},

$$(S_{11}-S_{12})_{\text{latt}}=1.088\times10^{14}\ ,$$

$$S_{44\ \text{latt}}=-0.363\times10^{14}\ .$$

By introducing $\gamma_\infty=-72$ corresponding to a Au^+ ion,[10] we get the values for the lattice contributions shown in Table I. Raghavan, Kaufmann, and Raghavan[11] have analyzed a large number of experimental static electric field gradients in noncubic metals and developed systematics between the lattice or ionic contribution to the field gradient and the local conduction-electron contribution. Applying these Raghavan systematics to the analysis of the $\overleftrightarrow{S}$ tensor components, Fischer *et al.*[12] found that $(S_{11}-S_{12})_{\text{ce}}$ must be of a different sign than $(S_{11}-S_{12})_{\text{latt}}$ and dominate it. Table I lists values of $(S_{11}-S_{12})_{\text{ce}}$ and $S_{44\text{ce}}$ with this method of analysis. We find the ratio of conduction-electron to lattice contributions to be consistent with the systematics of Raghavan, Kaufmann, and Raghavan for r_{12} and larger for r_{44}.

With the relative simplicity of the gold electronic structure, efforts can be made to understand the probable sources of the dynamic electric field gradients whose coupling tensor components we have measured.

TABLE I. Experimental results, point-charge theoretical results, and conduction-electron contributions for the $\overleftrightarrow{S}$ tensor components in gold. The product $|QS|$ is given in statvolts and the $\overleftrightarrow{S}$ tensor components in statvolts cm^{-2}. The values of electric quadrupole moment (Ref. 6) $Q=0.547$ b and antishielding factor (Ref. 10) $(1-\gamma_\infty)=73$ have been chosen.

$\lvert QS_{44}\rvert_{\text{meas}}=7.11\times10^{-9}$ (±4%)	$\lvert Q(S_{11}-S_{12})\rvert_{\text{meas}}=7.4\times10^{-9}$ (±10%)
$(1-\gamma_\infty)S_{44\ \text{latt}}=-2.65\times10^{15}$	$(1-\gamma_\infty)(S_{11}-S_{12})_{\text{latt}}=7.94\times10^{15}$
$S_{44}=13.0\times10^{15}$ (±4%)	$S_{11}-S_{12}=-13.6\times10^{15}$ (±10%)
$S_{44\ \text{ce}}=15.7\times10^{15}$ (±4%)	$(S_{11}-S_{12})_{\text{ce}}=-21.5\times10^{15}$ (±10%)
$S_{44\ \text{ce}}/(1-\gamma_\infty)S_{44\ \text{latt}}=r_{44}=-5.9$	$(S_{11}-S_{12})_{\text{ce}}/(1-\gamma_\infty)(S_{11}-S_{12})_{\text{latt}}=r_{12}=-2.7$

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